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LOCKHEED MARTIN

**CAVITATION BUBBLE
NUCLEATION BY ENERGETIC
PARTICLES**

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CAVITATION BUBBLE NUCLEATION BY ENERGETIC PARTICLES

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ABSTRACT

In the early sixties, experimental measurements using a bubble chamber confirmed quantitatively the thermal spike theory of bubble nucleation by energetic particles: the energy of the slow, heavy alpha decay recoils used in those experiments matched the calculated bubble nucleation energy to within a few percent. It was a triumph, but was soon to be followed by a puzzle. Within a couple of years, experiments on similar liquids, but well below their normal boiling points, placed under tensile stress showed that the calculated bubble nucleation energy was an order of magnitude less than the recoil energy.

Why should the theory work so well in the one case and so badly in the other? How did the liquid, or the recoil particle, "know" the difference between the two experiments? Another mathematical model of the same physical process, introduced in 1967, showed qualitatively why different analyses would be needed for liquids with high and low vapor pressures under positive or negative pressures. But, the quantitative agreement between the calculated nucleation energy and the recoil energy was still poor—the former being smaller by a factor of two to three.

In this report, the 1967 analysis is extended and refined: the qualitative understanding of the difference between positive and negative pressure nucleation, "boiling" and "cavitation" respectively, is retained, and agreement between the negative pressure calculated to be needed for nucleation and the energy calculated to be available is much improved. A plot of the calculated negative pressure needed to induce bubble formation against the measured value now has a slope of 1.0, although there is still considerable scatter in the individual points.

1. INTRODUCTION

Under normal conditions, most liquids have rather well-defined freezing and boiling points. However, it is well known that clean liquids can be supercooled or superheated, finally changing phase only considerably below the freezing, or above the boiling, point. The superheating effect is easily demonstrated with a clean glass of previously boiled water and a microwave oven. However, I do not recommend the experiment, because the final boiling event is fairly violent: liquid water at atmospheric pressure and, say, 125°C contains a lot of stored energy—almost one-fifth of that stored as chemical energy in the same volume of gunpowder, for example, and enough to flash about 5% of its mass into steam immediately, increasing the volume of that 5% more than a thousandfold.

Superheating is put to good use in the bubble chamber, invented by Donald Glaser and for which he was awarded the Nobel prize for physics in 1960. His first demonstration of particle tracks, in the early 1950s, involved a small glass bulb on the end of a long tube (Glaser 1958 and 1964). The apparatus was filled with diethyl ether, and the few mL in the bulb part was kept at 135°C or above, the rest of the system being at room temperature. The normal boiling point of diethyl ether at atmospheric pressure is only 34°C, but a piston pressurized the system, keeping even the ether in the bulb above its saturation pressure of about 13 atm and offering it no temptation to boil. However, when the pressure was released by moving the piston with a hand crank, the ether in the bulb was more than 100°C above its boiling point. Even so, there was a significant delay before general boiling occurred, and during that period, atomic particles (e.g., cosmic rays) passing through the bulb could be seen, by high-speed photography, to leave a track of small, but growing, bubbles. Stereo photographs of the bulb allowed the particle's path to be reconstructed. It was like a Wilson cloud chamber, but with a medium whose density was the order of 1 kg/L instead of only 1 to 100 g/L, and therefore gave a much greater chance of interacting within the volume of the chamber with an energetic particle.

Progress was rapid. In less than seven years, bubble chambers had grown from Glaser's tiny demonstration to the status of major, high-tech hardware: Berkeley had a 6-ft bubble chamber filled with 500 L of liquid hydrogen!

All of these machines worked on the same principle: the pressure on a suitable liquid was dropped suddenly from above the saturation value to below it, so that bulk boiling became energetically favorable. In so called "clean" bubble chambers like Glaser's first glass model, the liquid could remain in the superheated state for seconds, or longer, awaiting a particle whose path would then be outlined as a trail of bubbles. If the particle decayed, or interacted with an atom in the bubble chamber medium, the paths of the reaction products would also be rendered visible.

"Dirty" bubble chambers, including all of the biggest chambers, were those with structures or materials (such as seals and fasteners around the windows, and the piston) that would always nucleate boiling—like the rough, porous ceramic disks that used to be placed in kettles and pans to prevent the "bumping" that was, in fact, superheating and subsequent violent boiling—as soon as the pressure was dropped. The bubbles nucleated in that way eventually would repressurize the system to somewhere close to the saturation pressure, thus removing the superheat and rendering the chamber insensitive to incoming particles. However, the growth rate of the bubbles was slow enough that there was still a period of sensitivity lasting at least several milliseconds, before the pressure became too high.

One major disadvantage of the bubble chamber was that it could not be triggered. In a cloud chamber, on the other hand, the droplets making up the tracks in a supercooled, and therefore supersaturated, gas/vapor mixture are nucleated along the line of electric charges left by the incoming particle. Those charges remain separated long enough that a cloud chamber can be depressurized after the incoming particle has arrived and still show tracks. This means that interesting events can be identified by electronic detectors, such as Geiger or scintillation counters, placed outside the chamber in various coincidence/anticoincidence arrangements, and depressurization can be triggered if, but only if, such an event is detected.

No one ever succeeded in triggering a bubble track chamber in that way, or even in measuring the lifetime of whatever nucleation centers are left in a subcooled (i.e., pressurized) bubble chamber liquid by the incoming particles. Glaser and Rahm (1955) showed that the lifetime of the radiation-induced nucleation centers in a bubble chamber (meaning the length of time that a nucleation center created under pressure and temperature conditions unfavorable to growth, which Glaser referred to as a "latent image," remains capable of forming a bubble if conditions later become favorable) was not more than 1 ms, or in some cases less than 0.1 ms. They argued that it might be less than a few microseconds. West and Howlett (1968) reported experiments with very rapidly (ultrasonically) depressurized liquids that showed the lifetime of latent nucleation centers to be less than 2 μ s.

Some experiments, in which a small, clean, isopentane bubble chamber was placed in a waveguide where it could be zapped with a 2- μ s microwave pulse from a 250-kW magnetron (Bertanza et al. 1956) reported that boiling could be triggered after a particle had passed through. The chamber was depressurized, but not enough to show tracks; as a clean chamber, it could remain in this state for a significant period. During that period, a microwave pulse was initiated on detection of coincident signals in two Geiger-Muller (G.M.) counters, one above and one below the chamber, indicating that a particle had passed through the superheated isopentane. "In a certain range of temperature (from 80°C upwards) microwaves in coincidences with G.M. counters *always* give rise to boiling, whereas microwaves alone *never* do (of course, counter coincidences alone do not give rise to any boiling). . . ." Without knowledge of the delays in the electronics and of the microwave pulse rise time, which the reference does not supply, one cannot estimate the lifetime of the nucleation centers between their creation by the particle and their growth beginning during the microwave pulse. Furthermore, there are some puzzling aspects to the results (e.g., the bubbles photographed in the boiling liquid were few and scattered—no tracks were seen.

Years after bubble chambers were in common use, debate on the bubble nucleation mechanism continued. Glaser originally thought that the mechanism might be similar to the cloud chamber, depending upon ionization and electric charge effects, and others pursued the same idea. Eventually, a different theory gained general acceptance; its most eloquent and influential expression is probably found in a paper written by Seitz (1958), although by that time Glaser too had arrived at a similar viewpoint. In Seitz' theory, kinetic energy is transferred to the atoms and molecules of the medium, thus raising the temperature locally, producing "... highly localized hot regions, or 'temperature spikes,' within the liquid that literally explode into bubbles of larger than critical size, which can grow through evaporation of the superheated liquid." There is a critical size, beyond which the bubble can grow indefinitely, because the surface tension, σ , of the liquid effectively acts as an additional pressure, equal to $2\sigma/r$, tending to collapse the bubble. If the difference between the vapor pressure inside the bubble and the pressure outside it is greater than $2\sigma/r$, the bubble can continue to grow; i.e., the condition for growth is

$$P_{vap} > P_{ext} + 2\sigma/r ,$$

where P_{ext} is the pressure in the liquid after the chamber has been depressurized. The critical radius is therefore

$$r_{crit} = \frac{2\sigma}{P_{vap} - P_{ext}} \quad (1)$$

Seitz also calculated that the heat within the spherical thermal spike dissipated by conduction, losing all but a fraction $1/e$ of the initial heat content in a time τ , called the relaxation time, determined by the radius of the spike and the thermal diffusivity of the medium

$$\tau = r^2/4D \quad . \quad (2)$$

In a typical calculation for a bubble chamber liquid (hydrogen or propane) sensitive to minimum ionization particles, τ is the order of 10^{-11} to 10^{-10} s for a spike of the critical radius defined by Eq. (1).

Such a time was much too short for it to be practical to heat or depressurize the liquid chamber after particles were detected by external counters (i.e., there seemed to be no way in which a bubble chamber could be triggered by thermal or mechanical means). However, Rice-Evans (1973) proposed applying a fast-rising, high-gradient electric field to an already superheated liquid. The principle was that although the thermal spike would have dissipated before the pulse was applied, electrons and ions along the particle track would remain separated for longer and could be accelerated by the field, causing very localized heating and bubble nucleation. As far as I know, no such chamber was built, unless the Bertanza et al. results were due to an effect of this kind.

2. NEGATIVE PRESSURES

In 1956, Pless and Plano reported the successful construction and operation of a small, "clean" isopentane bubble chamber in which the liquid was actually placed under a negative pressure (i.e., tensile stress) in the expansion process.

The idea of a negative pressure is a little strange at first, but it is an entirely respectable, and quite common, phenomenon (e.g., Donny 1846; Berthelot 1850; Worthington 1892). For example, at the base of a tree, the sap is at approximately atmospheric pressure (a cut does not cause sap to spurt out or air to be sucked in). At the top of, say, a hundred-foot tree, the sap is at almost -2 atm because the pressure in a column of liquid water falls by 1 atm for every 34 ft of vertical height.

I can almost feel the readers' skepticism, but in fact there is a simple, convincing demonstration (Scholander et al. 1965). From the ground, a marksman with a rifle shoots through a twig near the top of a tall tree. The severed twig falls to the ground and, upon examination, it is found that the sap has retracted from the cut some distance into the capillaries. The twig is now placed into a pressure chamber with the cut end protruding, through a cuff seal, into the air. The pressure in the chamber is increased, and sap is forced out of the twig. The gauge pressure in the chamber needed to bring the surface of the sap level with the cut end is the same as the negative pressure that was in the liquid-filled capillaries of the twig at the instant that it was severed.

Evidently the capillaries of a tree are "clean" in the same sense as Glaser's first bubble chamber [i.e., their surface does not nucleate bubbles of vapor or of dissolved air in the sap, except under extreme conditions of negative pressure due to rapid loss of water from the leaves (Milburn and Johnson 1966)]. Likewise, Pless and Plano (1956) found that isopentane "will not boil from clean glass surfaces, even under a negative pressure as great as 6 atm. However, it boils quite readily from brass. Ethylene glycol ($C_6H_6O_2$), on the other hand, does not boil from brass under these same conditions." They devised an apparatus in which isopentane was in contact only with glass, while only ethylene glycol, which is immiscible with isopentane, came in contact with the brass and rubber of the moving piston device that was used to depressurize the system. They were able to produce negative pressures in excess of 4 atm, while the isopentane, at 120°C , had a vapor pressure of about 11 atm so that $P_{\text{vap}} - P_{\text{ext}} \approx 15$ atm. Incidentally, in their 1956 paper Pless and Plano also supported a thermal, rather than electric, charge basis for bubble nucleation.

B. Hahn (1961) measured the threshold (negative pressure) for nucleation of bubbles by α -decay recoils, fast neutrons, and gamma rays in various liquids under negative pressures. He achieved the negative pressures using a modification of a very simple method first, I think, used by Reynolds (Worthington 1892) and improved by Briggs (1949). In Hahn's experiments, a glass capillary tube with bent-back ends was rapidly rotated about an axis through its center (Fig. 1). The pressure over the free surface of the liquid near the ends of the capillary tube is atmospheric, but there is a pressure gradient along the tube, this time a much steeper one than in our tree because the radial acceleration in a centrifuge at high rotational speeds is much greater than the acceleration due to earth's gravity. In some liquids, negative pressures of more than 100 atm can easily be produced near the center of the tube. Hahn found that recoils from the decay of α -emitters dissolved in the liquid under test were very effective at nucleating bubbles, and even at room temperature the threshold in isopentane was only -3.3 bars.

Fig. 1. Rotating pyrex glass capillary tube (Hahn 1961).

Another way of producing negative pressures in a liquid is to subject it to high-intensity sound waves. It is not difficult to achieve sound pressure amplitudes of several atmospheres in some liquids, and such an amplitude imposed on a static pressure of, say, 1 atm will put the liquid under negative pressure during most of the negative-going half of the cycle.

In several experiments (Lieberman 1959; Hughes 1960; Sette and Wanderlingh 1962; Hahn and Peacock 1963), evidence was found of bubble formation, nucleated by energetic particles, in ultrasonically excited liquids. Sette and Wanderlingh's experiments showed that neutron irradiation could lower the cavitation threshold in water for several hours after the irradiation took place—this is in the strongest possible contrast to the no-memory behavior of bubble chambers. As far as I know, the nature of Sette's long-lived, radiation-induced nucleation centers has never been definitely identified, although the effect was verified in experiments by Finch (1964).

A dictionary definition of cavitation (below) is bubble formation under reduced pressures, which would not differentiate between, for example, the ultrasonic cavitation bubbles mentioned in the last paragraph and the boiling of liquid hydrogen in the Berkeley bubble chamber.

Webster's Third New International Dictionary

Cavitation . . . 1a: the formation of partial vacuums in a liquid esp. as a result of the passage through it of a swiftly moving solid body (as a propeller blade) or of high frequency soundwaves

1b: the pitting and wearing away of solid surfaces (as metal or concrete) as a result of the collapse of those vacuums

I do not know that there is any formal demarcation between the phenomena of boiling and cavitation, but I believe that many workers in the field have a sense that "cavitation" refers to "cold" liquids, those at such a temperature that the vapor pressure is small compared with the absolute value of the external pressure. In this sense, Pless and Plano's negative pressure isopentane bubble chamber at 120°C ($P_{vap} \approx 11$ bars, $\Delta P \approx 4$ bars) is in the boiling category, but Hahn's spinner measurements on the same liquid at 20°C ($P_{vap} \approx 0.8$ bars, $\Delta P \approx 3.3$ bars) represent cavitation. In a later section, a boundary line between the two categories of radiation-induced bubble formation will be quantified and seen to have some physical significance.

It should be noted that in other kinds of experiments, cavitation bubbles may contain significant amounts of permanent gas, although in this report I am not addressing that situation.

3. EXPERIMENTAL MEASUREMENTS

G. Riepe (then a graduate student) and B. Hahn carried out a wonderful series of experiments with a temperature-controlled bubble chamber specially designed to hold the pressure nearly constant for a long period (up to 100 ms) following the expansion. Using two different liquids, Freon 12 at several different temperatures between 30°C and 80°C and propane at 18.5°C, they measured the pressure drop necessary to nucleate bubbles by individual recoils from the α -decay of $^{210}\text{Polonium}$, and of $^{212}\text{Bismuth}$ and $^{212}\text{Polonium}$ (also known as Thorium C and Thorium C'). The results in Appendix A are measured from the graphs published in their paper (Riepe and Hahn 1961). Riepe and Hahn also measured the threshold for bubble formation by dissolved ^{37}Ar , which decays and releases an Auger electron with an energy of 2.3 keV.

The alpha-decay results are probably the easiest to analyze, because all of the recoil energy, but very little of the alpha's energy, is deposited in a small region, so that the energy available for bubble nucleation may be reasonably easily, but not necessarily accurately, estimated.

El-Nagdi and Harris (1971) carried out another clever experiment. To permit a higher superheat in the bulk of the hot liquid in a dirty bubble chamber, they cooled the walls of the chamber just before depressurizing it. With correct timing, a layer of colder liquid could be created around the walls while an interior core of hot liquid remained in the middle of the chamber. They measured the threshold (i.e., the superheat at which boiling began) in liquids irradiated by fast neutrons from an accelerator source.

Dietrich and Cornolly (1973), using a small, specially built, water bubble chamber measured the superheat needed for uranium fission fragments to nucleate bubbles.

Apfel (1979) developed a very different form of the bubble chamber, a most elegant and interesting idea, specifically for use as a neutron dosimeter. In Apfel's Superheated Drop Detector, very many uniform, small ($\sim 100\text{ }\mu\text{m}$ to 1 mm) droplets of a liquid whose normal boiling point is below room temperature are suspended in a gel. At room temperature and pressure, each and every small drop is superheated and is a miniature, one-shot, clean bubble chamber. Apfel (1979) and Lo and Apfel (1988) reported on a series of experiments in which the superheated drops were irradiated with monochromatic—or nearly so—neutron beams of different energies, and the number of droplets that turned to vapor, per unit time and volume, was measured. In some cases, where only fast neutron knock-ons were believed capable of nucleating bubble formation, there was a fairly well defined threshold of neutron energy for droplet vaporization.

Besides Hahn's negative pressure results with the spinner, thresholds for prompt neutron-induced ultrasonic cavitation (i.e., cavitation not involving the unexplained delayed effect reported by Sette and Wanderlingh and by Finch) were reported by Greenspan and Tscheig (1967).

For the convenience of future researchers in the field, most of the results from the above measurements, converted to a common set of units and presented in a common format, are collected in Appendix A.

4. RIEPE AND HAHN'S BUBBLE CHAMBER MEASUREMENTS AND THEORY

As described above, Riepe and Hahn measured the threshold for bubble formation by alpha-recoils and Auger electrons in a bubble chamber specially designed to have a long (~100 ms) period of constant, reduced pressure following the rapid (~4 ms) expansion.

They compared the alpha-decay results with calculations based on Seitz' theory. A basis of their calculation is that a static bubble with internal pressure P_{vr} will tend to grow if its radius is r_{crit} or greater where

$$r_{crit} = \frac{2\sigma}{P_{vr} - P_{ext}} \quad (3)$$

This is similar to Eq. (1), except that they applied a correction to the vapor pressure inside the bubble to account for the curvature of the bubble surface. Now, it is known that the equilibrium vapor pressure over a tiny droplet is significantly higher than over a plane surface (see Wilson 1951), but Glaser (1958) believed that there was no corresponding effect in bubbles (i.e., no decrease in equilibrium vapor pressure within the concave liquid surface of a small bubble, although Peyrou (1967) argued for such a decrease). In any case, Riepe and Hahn incorporated such an effect into their bubble calculations, hence the appearance of P_{vr} instead of P_{vap} in Eq. (3). Their formula for the ratio of the equilibrium vapor pressure over a plane surface, P_{vap} , to that over the surface of radius r_{crit} , P_{vr} , was

$$\ln (P_{vap}/P_{vr}) = \frac{Mwt}{RT\rho_\ell} \cdot \frac{2\sigma}{r_{crit}} \quad (4)$$

R is the gas constant; ρ_ℓ is the density of the liquid (mass/unit volume). Mwt , σ , and T are its molecular weight, surface tension, and temperature, respectively. The calculations in this report use molar, rather than mass, densities, and so the multiplier Mwt is not needed here.

The curvature of a small droplet also changes the enthalpy of evaporation, but the effect is very small in the circumstances considered here (see Appendix B) and has been ignored in these calculations.

The measurements were made by expanding the chamber so that the pressure in the liquid, which contained traces of dissolved alpha-emitter or ^{37}Ar , fell from its equilibrium vapor pressure, P_{vap} , to some lower pressure, P_{ext} . For small pressure reductions, with a dissolved alpha-emitter, few or no bubbles were seen, but once the $\Delta P = P_{vap} - P_{ext}$ exceeded a threshold value, many bubbles were observed in the photographs of the chamber. At any particular ΔP above the threshold, the number of bubbles in the pictures was linearly proportional to the length of time for which the depressurization lasted. Increasing the ΔP slightly beyond the threshold gave more bubbles per picture, but further increases of ΔP beyond about 1.1 times the threshold brought no additional bubbles: apparently a ΔP only 10% or so above the threshold was enough to give a bubble for every single alpha-decay taking place in the sensitive region during the depressurized period.

Following Seitz, Riepe and Hahn calculated five different terms in the energy of formation of a critical bubble [Eqs. (5a-5e)]:

$$W_{sl} = \text{surface energy} = 4 \pi r_{crit}^2 \sigma \quad (5a)$$

$$W_e = \text{PV work done by the bubble during expansion to the critical size} = \frac{4}{3} \pi r_{crit}^3 P_{ext} \quad (5b)$$

$$W_v = \text{enthalpy of evaporation of the vapor filling the bubble} = \frac{4}{3} \pi r_{crit}^3 \rho_v H_v \quad (5c)$$

$$W_4 = \text{kinetic energy given to the liquid moving away as the bubble expands to the critical radius} = 32 \pi D^2 \rho_l r_{crit} \times (\text{Mwt}/10^3)^a \quad (5d)$$

$$W_5 = \text{viscous losses in moving away the liquid as the bubble expands} = 64 \pi \eta D r_{crit} \quad (5e)$$

^aThe term $\text{Mwt}/10^3$ is the mass, in kg, of 1 mol; it is needed to convert the density, ρ_l , from mols/m^3 to kg/m^3 so that the kinetic energy appears in units of joules. This factor was not needed in Riepe and Hahn's paper because they did not use molar enthalpies, densities, specific heats, etc.

For convenience later, I have partially rewritten the Riepe and Hahn equations to use some of Peyrou's (1967) notation. D is the thermal diffusivity; H is the enthalpy of evaporation; and η is the viscosity of the liquid.

The dynamic terms, W_4 and W_5 , were calculated on the assumption that the bubble expands at constant velocity to the critical radius in a time equal to the relaxation time τ for conductive dissipation of the heat from a spherical thermal spike of radius r_{crit} deposited in the bulk liquid.

$$\tau = r_{crit}^2 / 4D = r_{crit}^2 c \rho_l / 4\lambda \quad (6)$$

where D is the thermal diffusivity, $= \lambda / \rho_l c$, of a liquid of thermal conductivity λ , density ρ_l , and specific heat c .

The velocity v_c of the bubble wall is taken as the critical radius divided by τ ,

$$v_c \sim r_{crit} / \tau = 4D / r_{crit} \quad (7)$$

In comparing the above energy terms with the available energy, Riepe and Hahn assumed that all the energy deposited in a length equal to the diameter of the critical bubble was available. The alpha-recoils generally had a calculated range, R_r , less than $2r_{crit}$, and in that case the energy calculated to be lost by the alpha particle in a distance $2r_{crit} - R_r$ at the beginning of its range was added to the recoil energy. Two sets of results published by Riepe and Hahn are shown in Table 1. Appendices C and D recalculate these results using more recent values for the liquid properties, with the addition of another energy term proposed by Bugg (1959); the recalculations independently confirm the Riepe and Hahn conclusions.

The ratio of critical radius to velocity, in accordance with Eq. (7), reveals that the thermal relaxation time for these large thermal spikes is $\sim 10^{-8}$ s.

Table 1. Reipe and Hahn: Energy balance with recoils from ^{210}Po decay

	CCl_2F_2 at 29.4°C	C_3H_8 at 18.5°C
Threshold ΔP^a	2.00 atm	1.97 atm
Calculated critical radius r_c , b Å	860.8 Å	829.3 Å
Growth velocity (max)	3.04×10^2 cm/s	6.1×10^2 cm/s
No. of molecules in critical bubble	5.55×10^5	5.52×10^5
Surface energy, W_{sl}	4.9 keV	4.3 keV
PV work, W_e	8.7 keV	8.9 keV
Latent heat, W_v	94.7 keV	89.0 keV
Kinetic energy, W_4	0.3 keV	~0.4 keV
Viscous work, W_5	1.8 keV	~2.6 keV
Total energy needed	110.4 keV	105.2 keV
Recoil range R_r , c	651 Å	1146 Å
Recoil energy	101.0 keV	101.0 keV
Alpha-energy loss (in a distance $2r_{crit}-R_r$)	11.6 keV	2.8 keV
Total energy available	112.6 keV	103.8 keV

^aMeasured by CDW from the graphs in Fig. 6 and in Fig. 10, Riepe and Hahn (1961).

^bThis is the critical radius calculated by correcting the vapor pressure in the bubbles for the curvature effect.

^cFrom Table 1 of Riepe and Hahn; based on a formula privately communicated to them by Lindhard and Schiff.

The very close quantitative agreement between the calculations of energy needed and energy available for bubble nucleation by the heavy, slow α -recoils is impressive and confirms Seitz' basic theory. In practice, bubble chambers were more usually employed to detect minimum ionization particles, and in those circumstances there are a number of discrepancies between the theory and the observation: for a good discussion of those discrepancies, and some possible reasons, see Section IV in Peyrou (1967).

5. HAHN'S SPINNER RESULTS—A MAJOR PUZZLE

5.1 THE RESULTS

As we have seen, the thermal spike theory, as formulated by Seitz, gave a quantitatively accurate explanation of the nucleation of boiling by α -decay recoils in propane and Freon 12. But a surprising thing emerged when Hahn (1961) measured the negative pressure needed to form macroscopic bubbles in some other liquids at temperatures below their normal boiling points from these same particles and from fast neutrons. His 1961 paper simply reported the results from spinner experiments, but a later one (Hahn and Peacock, 1963) noted that in the negative pressure experiments, by both the spinner and ultrasonic methods, the calculated energy needed to form viable nucleation centers, typically ~ 17 keV for the static terms, was very much lower than the energy, ~ 100 keV, thought to be available within the critical diameter from the recoiling nuclei. With some understatement they wrote, "In contrast to the behavior at positive pressures, as in normal bubble chamber operation, where energy equilibrium was found with the above terms, at negative pressures additional terms seem to play an important role."

Table 2 lists the negative pressure thresholds measured by Hahn and Peacock for bubble formation by α -decay (mostly ^{210}Po decay) and some properties of the liquids. These negative pressures correspond to the value P_{ext} in Eqs. (3) and (5). Using the TK Solver models described in Appendix C, the parameters of critical bubbles under these conditions were calculated and are also listed in Table 2. Table 3 lists the liquid and vapor properties, from the Yaws (1996) data, needed to calculate the Seitz theory energy terms; Appendix E also lists those properties and the ones used, later in this report, for calculations based on a different model.

Table 2. Results from Hahn's spinner experiments (with ^{210}Po α -decay except as indicated)^a

Material	Composition	Temp., °C	Recoil energy (keV)	Measured P_{neg} (bars)	Vapor pressure [kPa (Yaws)]
Isopentane	C_5H_{12}	20	101	3.344	76.7
Freon 113 ^b	$\text{C}_2\text{Cl}_3\text{F}_3$	"	166	4.490	36.1
Freon 113	$\text{C}_2\text{Cl}_3\text{F}_3$	"	101	5.207	36.1
n-hexane	C_6H_{14}	"	"	6.890	16.3
Acetone	$\text{C}_3\text{H}_6\text{O}$	"	"	8.309	24.6
Trichloroethylene	C_2HCl_3	"	"	11.754	7.75
Ethylene bromide	$\text{C}_2\text{H}_4\text{Br}_2$	"	"	22.190	1.36
Acetophenone	$\text{C}_8\text{H}_8\text{O}$	20.5 ^c	"	33.235	0.037
Cyclohexanol	$\text{C}_6\text{H}_{12}\text{O}$	23.45 ^d	"	48.740	0.088

^aResults taken from Table A.2.

^bResult with Th (C + C') α ; measured from Fig. 2, Hahn 1961.

^cHahn's paper indicates that all these spinner experiments were carried out at 20°C, but the normal freezing point of acetophenone is 20.5°C.

^dHahn's paper indicates that all these spinner experiments were carried out at 20°C, but the normal freezing point of cyclohexanol is 23.45°C.

Table 3. Liquid and vapor properties for calculations of bubble formation energy in ^{210}Po experiments

Name	Material and conditions									
	Isopentane	Freon 113	n-hexane	Acetone	Trichloroethylene	Ethylene bromide	Acetophenone	Cyclohexanol		
Composition	C_5H_{12}	$\text{C}_2\text{Cl}_3\text{F}_3$	C_6H_{14}	$\text{C}_3\text{H}_6\text{O}$	C_2HCl_3	$\text{C}_2\text{H}_4\text{Br}_2$	$\text{C}_8\text{H}_8\text{O}$	$\text{C}_6\text{H}_{12}\text{O}$		
Molecular weight	72.150	187.375	86.177	58.080	131.388	187.862	120.151	100.161		
Temperature (T, °C)	20	20	20	20	20	20	20.5	23.45		
P_{ext} , ^a kPa	-334	-521	-689	-831	-1175	-2219	-3323	-4874		
Vapor properties										
Pressure P_{vapor} , kPa	76.69	36.10	16.32	24.58	7.75	1.36	.037	.088		
Density, ^b mol/m ³	31.97	14.70	6.51	9.82	3.06	0.515	.013	.029		
Liquid properties (Yaws)										
Surface tension, mN/m	14.99	17.72	18.54	23.66	29.40	38.82	39.46	33.58		
$T\partial\sigma/\partial T$, mN/m	-31.73	-33.16	-32.56	-36.26	-38.88	-39.09	-33.09	-31.41		
Density, kmol/m ³	8.607	8.411	7.670	13.619	11.158	11.600	8.556	9.605		
Enthalpy of evap., kJ/mol	25.27	28.86	32.28	32.56	35.27	38.38	53.89	61.09		
Specific heat, J/mol · K	157.03	168.26	191.40	126.66	123.05	131.03	199.31	217.80		
Thermal conductivity, W/m · K	0.1156	0.771	0.1225	0.1630	0.1176	0.1013	0.1742	0.1343		
Viscosity, mPa · s	0.225	0.734	0.310	0.322	0.571	1.749	1.891	27.54		
Thermal diffusivity	8.553×10^{-8}	5.448×10^{-8}	8.344×10^{-8}	9.449×10^{-8}	8.565×10^{-8}	6.665×10^{-8}	10.17×10^{-8}	6.420×10^{-8}		
Calculated values										
Alpha ^c	0.9878	0.9766	0.9646	0.9646	0.9580	0.9246	0.8529	0.8140		
r_{crit} , ^d kPa	73.17	63.71	52.62	55.36	49.73	34.97	23.75	13.78		
P_{vapor} , ^d kPa	75.75	35.254	15.73	23.71	7.424	1.257	0.0316	0.0716		
Vapor density, ^e mol/m ³	31.97	14.70	6.51	9.82	3.06	0.515	—	—		
P_{vapor}/RT , ^e mol/m ³	31.07	14.46	6.45	9.73	3.05	0.0516	0.013	0.029		

^aThe measured value of negative pressure threshold from Table A.2.

^bAt temperature T, pressure P_{vapor}

^cRatio of the vapor pressure over a concave surface of radius r_{crit} to the vapor pressure over a plane surface (see Appendix B).

^dAllowing for a decrease in vapor pressure over a concave surface.

^eThese two lines show the difference between the actual vapor density (Yaws) and the density of an ideal gas under the same conditions.

5.2 ENERGY BALANCE

Table 4 lists the energy terms for the negative pressure results in the same manner as the bubble chamber results in Table 1 but with Bugg's additional surface energy term (see Appendix D) also included. One sees that indeed the sums of all the energy terms are much less than the energy (~ 100 keV) of the α -recoil, even if the Bugg term is included. The relative contributions of the terms are also very different. For example, the enthalpy of vaporization is generally a minor, rather than a major, term. That is because in cavitating liquids the vapor pressure is, by my definition of cavitation, relatively small, and therefore, the mass of vapor in the critical bubble is small.

In some cases, the critical radius is much smaller than in the bubble chamber, because of the larger pressure difference between the interior of the bubble and the liquid outside it, so that we may imagine that for those liquids the entire recoil energy might not be available in a distance $2r_{crit}$. However, for isopentane and Freon 113, the critical radii are quite comparable with those in the Riepe and Hahn bubble chamber experiments, so that such an effect cannot be the whole explanation for the discrepancy between the calculated energy needed to form the nucleation center and the recoil energy.

Why does a theory that worked so splendidly for the bubble chamber alpha recoil experiments fail so badly when applied to the same particles in similar liquids at lower temperatures?

Table 4. Energy terms (in keV) for Hahn's spinner liquids nucleated by ^{210}Po recoils

Term	Isopentane	Freon 113	n-hexane	Acetone	Trichloroethylene	Ethylene bromide	Acetophenone	Cyclohexanol
W_{s1}	6.3	5.6	4.0	5.7	5.7	3.7	1.7	0.5
W_c	3.4	3.5	2.6	3.7	3.8	2.5	1.2	0.3
W_v	8.3	2.9	0.8	1.4	0.3	~ 0.0	~ 0.0	~ 0.0
Subtotal ^a	18.0	12.0	7.4	10.8	9.8	6.2	2.9	0.8
W_{s2}	13.3	10.6	7.1	8.7	7.5	3.7	1.5	0.5
Subtotal static terms ^b	31.3	22.6	14.5	19.5	17.3	9.9	4.4	1.3
W_4	0.2	0.2	0.2	0.4	0.3	0.2	0.2	~ 0
W_5	1.8	3.2	1.7	3.1	3.1	5.1	5.8	30.6
Total ^c	33.3	26.0	16.4	23.0	20.7	15.2	10.4	31.9

^aRiepe and Hahn found that these static terms alone provided a good energy balance for their bubble chamber experiments.

^bStatic terms including Bugg's additional surface energy term.

^cStatic and dynamic terms.

6. CAVITATION BUBBLE NUCLEATION

6.1 CALCULATION OF MINIMUM mT

A possible explanation for the difference between boiling and cavitation nucleation events was proposed a few years later (West 1967). It is based on the fact that the Seitz condition ($P_{vr} - P_{ext} > 2\sigma/r_{crit}$) is necessary for growth to proceed from r_{crit} , but may not be sufficient to ensure growth up to that radius. Instead of the Seitz condition at the critical radius, the new model required that

$$P_{int}(r) - P_{ext} \geq 2\sigma/r \quad (8)$$

at all stages of growth from the initial "... highly localized hot regions ..." described by Seitz. $P_{int}(r)$ is the pressure in the bubble when its radius is r . For the negative pressure case, $P_{ext} = -P_{neg}$.

For simplicity, the vapor inside the bubble was treated as an ideal gas, although the last two lines of Table 3 show that it really is not. Then, during the growth

$$\frac{m(r)T(r)R}{V(r)} + P_{neg} \geq \frac{2\sigma}{r}$$

or

$$\frac{m(r)T(r)R}{(4/3)\pi r^3} + P_{neg} > \frac{2\sigma}{r}, \quad (9)$$

where $m(r)$ is the mass of gas inside the bubble, and $T(r)$ is its temperature, when the radius is r . Note that according to Eq. (9), there is one radius, $r^* = 2\sigma/P_{neg}$, beyond which the bubble would continue to grow even if there were no gas inside it at all.

As a gross oversimplification, suppose first that $m(r)T(r)$ remains constant during the growth. Then at a very small positive r , the first term will dominate in Eq. (9), thus satisfying the inequality. At a very large positive r , the first and third terms tend to zero, the second term dominates, and again the inequality is satisfied. Figure 2 illustrates this by plotting the left hand side (LHS) and right hand side (RHS) of Eq. (8). Two cases are shown: one in which P_{neg} ($= -P_{ext}$) is large enough that the inequality is maintained for all r and one in which P_{neg} is such that, although the inequality is fulfilled at large and small r , there is an intermediate region in which it is not and through which the bubble could therefore not grow.

If mT is a constant, the condition that the curves never cross is that the simple cubic equation in $1/r$,

$$\frac{3mTR}{4\pi} \cdot \left(\frac{1}{r}\right)^3 - 2\sigma \left(\frac{1}{r}\right) + P_{neg} = 0$$

Fig. 2. The expansive (LHS) and compressive (RHS) pressures on bubbles filled with a constant (mass $\times$ temperature) of vapor.

or

$$\left(\frac{1}{r}\right)^3 - \frac{8\pi\sigma}{3mTR} \left(\frac{1}{r}\right) + \frac{4\pi P_{neg}}{3mTR} = 0, \quad (10)$$

does not have the two real, positive roots represented by the two crossing points of the curves in Fig. 2, Example 2. The equation is a reduced cubic (i.e., it is of the form $y^3 + py + q = 0$). The condition that the two real roots are equal, so that the curves touch but do not cross, can be found in older algebra text books; it is that $(p/3)^3 + (q/2)^2 = 0$. If $(p/3)^3 + (q/2)^2 > 0$, the curves in Fig. 2 do not even touch, and the internal pressure always exceeds the sum of the external pressure and the surface tension effect, $2\sigma/r$. Substituting the coefficients p and q from Eq. (10) provides the condition that the compressive pressures on the bubble never exceed the expansive ones,

$$mT \geq \frac{128\pi\sigma^3}{81RP_{neg}^2}. \quad (11)$$

The threshold (i.e., the minimum negative pressure to cause cavitation) would be given by choosing the equality sign, when

$$P_{neg} = \sqrt{\frac{128\pi\sigma^3}{81mTR}}. \quad (12)$$

In Eq. (11), if the equality sign (i.e., the limiting case or threshold) is chosen, then there is one positive radius, and one only, at which the expansive and compressive pressures in Eq. (8) are equal, while at all other real, positive radii, larger or smaller, the expansive pressure is the greater. Cardan's solution to a cubic equation (see Korn and Korn 1968) shows that the radius, r_{eq} , at which the two are equal is given by

$$r_{eq} = \sqrt[3]{\frac{2}{q}} = \frac{4\sigma}{3P_{neg}}. \quad (13)$$

The pressure, P_{int} , of the ideal gas inside the bubble at that time is equal to mTR divided by its volume,

$$P_{int} = \frac{mTR}{(4/3)\pi r_{eq}^3}. \quad (14)$$

Substituting for mT from Eq. (11) and for r_{eq} from Eq. (13) yields an expression for the pressure inside the bubble at the equilibrium radius,

$$P_{int}(r_{eq}) = P_{neg}/2, \quad (15)$$

a delightfully simple result.

It is interesting, and reassuring, that subsequent—but completely independent—research into the effects of helium generated by transmutation in irradiated solids led to equations that are formally identical to the original mT model of bubbles in liquids. For example, compare Eqs. (13) and (12) above with Eqs. (3) and (10), and Eq. (11), in Mansur et al. (1986).

6.2 BOILING AND CAVITATION

Now one can see a possible difference between boiling and cavitation bubbles. If the vapor pressure is low (cavitation case), Eq. (11) must be fulfilled to ensure growth from Seitz' initial small bubble, through the most difficult size (radius around r_{eq}). But, if the vapor pressure is high (boiling case) and evaporation from the bubble surface prevents the internal pressure from falling below P_{vap} , the LHS of Eq. (9) would look like the curve in the second example of Fig. 2 only until the pressure fell to P_{vap} , when the LHS curve would flatten out, remaining above the RHS curve and thus ensuring continued growth, as shown in Fig. 3, which is the same as Fig. 2, second example, except that the vapor pressure, which was much less than $P_{neg}/2$ in Fig. 2, is now set to $P_{vap} = 3 \times P_{neg}$. Physically, what has been done in Fig. 3 is to maintain $m(r)T(r)$ constant only until $P(r)$ falls to P_{vap} , at which time evaporation from the bubble surface adds more vapor, increasing m enough to keep the pressure constant. The effect is to hold the internal pressure higher than $P_{neg}/2$, the minimum equilibrium pressure at the most difficult stage of growth around r_{eq} .

Obviously, this is a grossly oversimplified model, but it demonstrates why there might be a qualitative difference in the nucleation energetics of boiling and cavitation. From this model, one would expect that in any negative-pressure experiment showing a negative-pressure threshold for bubble

Fig. 3. Expansive and compressive pressures when P_{vap} is three times P_{neg} (small P_{neg} , large P_{vap}).

nucleation of less than twice the vapor pressure, and in all positive pressure experiments, the Seitz and the Riepe and Hahn approaches based on vapor pressure would be applicable. The model described in this section would be needed to explain the experiments showing a negative-pressure threshold of more than twice the vapor pressure, which, as seen in Table 2, include all of Hahn's reported spinner results.

6.3 INITIATION OF BUBBLE FORMATION IN COLD LIQUIDS

Measurements of the superheat limit (i.e., the maximum temperature to which a liquid can be taken before vapor bubbles are spontaneously nucleated) are usually made by allowing a droplet of the test liquid to rise slowly through a bath of another, immiscible liquid with higher density and a higher boiling point, such as glycerin or sulfuric acid (see Apfel 1972 or Carey 1992).

A temperature gradient is maintained in the bath so that the temperature at the bottom is lower than, and at the top is much higher than, the boiling point of the liquid under test.

The droplets are small enough, typically ~ 1 mm diam, and purified enough that the probability of an inhomogeneous nucleation center, such as a speck of dust, being found in any particular droplet is low. They are also small enough, and rise slowly enough, to be in equilibrium with the temperature of the liquid immediately surrounding them. Observing the position at which the rising droplets explode into vapor, and knowing the bath temperature as a function of depth, one can find the superheat at which homogeneous nucleation occurred. By superimposing an acoustic standing wave on the static pressure in the bath, the method can be extended to negative pressures (Apfel 1971).

Experiments of this kind reveal that a typical positive pressure superheat limit for organic liquids is in the range of $0.85T_{crit} - 0.9T_{crit}$, where T_{crit} is the critical temperature of the substance (Carey 1992). This is the case over a wide range of pressures, up to 10% or more of the critical pressure.

For high sensitivity, bubble chambers are normally operated 1/2 to 2/3 of the way between the boiling point and the critical point: at a higher temperature—called the foam limit in bubble chamber jargon—spontaneous or homogeneous nucleation fills the chamber with bubbles upon depressurization, whether any radiation traverses it or not.

Under bubble chamber conditions, the enthalpy of evaporation is more than the enthalpy required to raise the temperature of the liquid in Seitz' thermal spike from the operating temperature to $0.9 T_{crit}$, so that if enough energy is deposited to form the vapor in a bubble of critical size, we may think that the liquid in the spike is raised above the superheat limit and will indeed boil. This is shown in Table 5.

However, homogeneous nucleation is a statistical process, originating in the density fluctuations that arise and disappear over a scale a few molecules across. The typical droplet, which might contain 10^{19} – 10^{20} molecules and whose temperature is slowly varying (increasing) at a rate ~ 1 deg/s, will have a high probability of spontaneous nucleation before the superheat threshold is exceeded by more than a fraction of a degree. The Seitz thermal spike on the other hand contains only 10^5 – 10^6 molecules (see Table 1), and in order to have a high probability of spontaneous boiling within the 10^{-8} s or so before the heat is dissipated into the surrounding liquid, the initial temperature must be not just within 10% of the critical temperature, as is the case for the slowly heated macroscopic drops, but much closer. Therefore, I now assume that, in practice, the thermal spike must both be of greater than a critical size, calculated below, and heated to the critical point.

In the case of "cold" (i.e., cavitating) liquids, the enthalpy needed to raise the liquid temperature from the operating point to the critical point is actually greater than the enthalpy of evaporation (see Table 6). The amount of energy needed to promote a high probability of nucleation within the volume of the thermal spike is therefore sufficient to evaporate that entire volume of liquid.

Table 5. Operating temperatures, superheat limits, and enthalpies in Riepe and Hahn bubble chamber experiments

Material	Composition	Operating temp., T_{op} , K	Superheat limit $0.9 T_{crit}$, K	Enthalpies, kJ/mol	
				H_{vap}	$H(0.9T_{crit}) - H(T_{op})$
Freon 12	CCl_2F_2	303	347	16.5	5.7
Propane	C_3H_8	292	333	15.2	5.1

Table 6. Operating temperatures, critical temperatures, and enthalpies in Hahn's negative pressure spinner experiments

Material	Composition	Operating temp., T_{op} , K	Critical temp., T_{crit} , K	Enthalpies, kJ/mol	
				H_{vap}	$H(T_{crit}) - H(T_{op})$
Freon 113	$\text{C}_2\text{Cl}_3\text{F}_3$	293	487	28.9	45.0
Acetophenone	$\text{C}_3\text{H}_8\text{O}$	294	701	53.9	112.6

7. EXTENSIONS OF mT THEORY

7.1 HEATED MASS

The above observation suggests a way to estimate the initial value of mT in Eq. (9) or Eq. (11). If E_{av} , the energy available for bubble nucleation, is known, one can calculate the number of moles of liquid, M_o , initially at T_{amb} , that could be raised to the critical point, or very close to it. Within that heated fluid, homogeneous nucleation would take place, resulting in its entire mass turning into a bubble of heated vapor,

$$M_o = E_{av} / H_{ca} \quad , \quad (16)$$

where H_{ca} is the difference in enthalpy per mole between the liquid at T_{amb} and at T_{crit} . The value of H is not very dependent on pressure, and I simply used the difference between the values at T_{amb} , 1 atm, and T_{crit} , P_{crit} in making this calculation (a more exact course would be to take the enthalpy difference between P_{neg} , T_{amb} , and the critical point, but enthalpy data for negative pressure conditions were not available).

Some sophisticated simulations of interface and bubble formation in small volumes of liquid heated to the critical point by radiation are reported by Sun, Chu, and Apfel (1992 a and b). Their simulations include many more factors than Eq. (16), and are much more rigorous, realistic, and complete models, capable of providing insights and predictions concerning many aspects of the early stages of bubble formation and the fate of the energy deposited by the incoming particle. In this report, I have chosen to simplify, or oversimplify, the physical model so that general algebraic solutions can be found.

For the present, assume that all of the recoil energy is available, but neglect any contribution from the alpha particle (i.e., write $E_{av} = E_{recoil}$). Also, consider only static energy terms (but remember that Table 4 shows that the dynamic terms would be very important for cyclohexanol).

With these assumptions, it is easy to calculate the threshold negative pressure for alpha-recoil-induced cavitation by the mT method and by using the Riepe and Hahn approach.

However, a modification of the model used by West (1967) is introduced here to reflect the fact that close to the critical point, as the initial mass of heated fluid is in this new version of the mT model, the ratio of mass to volume (i.e., the density) can be very different from the ideal gas value of P/RT . Accordingly in Eq. (9), (10), and therefore (12), the value of RT is replaced by P_{crit}/ρ_{crit} . Equation (12) for the threshold pressure becomes

$$P_{neg} = \sqrt{\frac{128\pi\sigma^3 H_{ca}}{81 E_{recoil} P_{crit} / \rho_{crit}}} \quad (17a)$$

Since, from Eq. (16) $E_{recoil}/H_{ca} = M_o$, the mass of fluid heated to the critical point, Eq. (17a) can be rewritten as

$$P_{neg} = \sqrt{128\pi\sigma^3 / 81 P_{crit} V_o} \quad , \quad (17b)$$

where $V_o = M_o / \rho_{crit}$ is the volume of the mass M_o at the critical pressure and temperature. It is reassuring to note that if the fluid were to behave as an ideal gas, even at the critical point, Eqs. (17b) and (12) would be identical.

It is easy to show that the two equal real roots of the equation, where the surface tension forces are in equilibrium with the internal pressure and the external, negative pressure, are given by

$$r = r_{eq} = \frac{4\sigma}{3P_{neg}},$$

just as in Eq. (13).

In order to calculate the threshold from the Seitz/Riepe model, one needs to know the vapor pressure. Because the vapor pressure is relatively low in the case of cavitation, according to my definition, in these particular calculations I have neglected its small dependence on bubble radius.

The results from the mT and Seitz/Riepe models are compared in Table 7.

Inspection of Table 7 shows that Eq. (17), based on the simple mT model, follows the trend of the experimental results reasonably well, although the predicted thresholds are consistently low by a factor of about 1.5 to 2.8 (excluding the very high viscosity cyclohexanol, since only static terms were included in the calculations). The vapor pressure model is much less consistent, with the predicted thresholds low by a factor of 3–15 and not even in the same rank order as the measured values. This observation is quantified by making a least squares fit of the measured values and the two sets of calculated data (excluding the cyclohexanol results). See Figs. 4 and 5. The R^2 correlation coefficients are vastly

Table 7. Calculated and measured thresholds

Material	H_{ca} kJ/mol	Recoil energy, keV	Calculated threshold (bars) for different models		Measured threshold (bars)
			Seitz/Riepe ^a	Eq. (17)	
Isopentane	39.78	101	1.13	1.99	3.34
Freon 113	44.97	166	1.20	2.05	4.49
Freon 113	44.97	101	1.49	2.63	5.21
n-hexane	59.25	101	1.43	3.22	6.89
Acetone	39.87	101	2.26	4.06	8.31
Trichloroethylene	47.07	101	2.22	5.40	11.75
Ethylene bromide	63.88	101	2.64	8.94	22.19
Acetophenone	112.61	101	2.53	12.13	22.23
Cyclohexanol ^b	99.95	101	2.00	9.81	48.74

^aUsing only the three static energy terms (W_{st} , W_e , W_v) of the Riepe and Hahn bubble chamber calculations.

^bThis liquid has a very high viscosity compared with the others.

Fig. 4. $P_{neg}(exptl)$ vs $P_{neg}(calc, \text{vapor model})$. Static terms only. Cyclohexonal excluded. $R^2 = 0.658$.

Fig. 5. $P_{neg}(exptl)$ vs $P_{neg}[calc, mT \text{ model, Eq. (17)}]$. Static terms only. Cyclohexonal excluded. $R^2 = 0.993$.

different at 0.993 for the mT calculations, but only 0.658 for the vapor pressure-based results. This seems to indicate that the Seitz thermal nucleation model can be applied in all cases, but the algebraic form of it that explains Riepe's positive pressure bubble chamber measurements is indeed not applicable to the different physical circumstances of Hahn's cavitation experiments.

7.2 VARIABLE mT

In fact, the simplifying assumption in R-5486 (West 1967) that mT remains constant during the initial expansion of the nucleation center is inconsistent with the physical model being used. For the ideal gas considered in the basic mT model, a constant value of mT implies a constant internal energy. But, the growth is pictured as an adiabatic process, which means that the difference between the additional surface energy and the PV work done by the external, negative pressure during growth must be drawn from the internal energy of the gas within the bubble, thereby reducing mT as the expansion proceeds. Fortunately, especially when only these static terms are being considered, the mathematical expression of the mT model can be readily modified to accommodate an approximate representation of this effect.

In general, internal energy is equal to enthalpy minus PV, so the initial internal energy, U_o , of the nucleation center is given by

$$U_o = E_{recoil} - P_{crit} V_o \quad (18)$$

At a stage during the expansion when the bubble radius is r and its volume $V(r) = (4/3)\pi r^3$, the internal energy will have fallen by an amount equal to the work done on the growing bubble. So

$$U(r) = U_o - (W_{s1} + W_{s2}) - W_e \quad (19)$$

$$= U_o - 4\pi r^2 \sigma_{eff} + P_{neg} V(r) \quad (20)$$

where only static terms are included and $\sigma_{eff} = \sigma - T\partial\sigma/\partial T$ to include the additional surface energy term proposed by Bugg. If the Bugg term is not applicable, one would simply write $\sigma_{eff} = \sigma$.

For an ideal gas, pressure is proportional to the internal energy per unit volume, so

$$P_{int}(r) = P_{crit} \cdot \frac{U(r)/V(r)}{U_o/V_o} \quad (21)$$

Now substitute for $U(r)$ from Eq. (20),

$$P_{int}(r) = \frac{P_{crit} V_o}{U_o V(r)} \cdot [U_o - 4\pi r^2 \sigma_{eff} + P_{neg} V(r)] \quad (22)$$

Using this value for $P(r)$ in Eq. (8) and substituting $(4/3)\pi r^3 = V(r)$ yields

$$\frac{P_{crit} V_o}{U_o} \left(\frac{3}{4\pi} \cdot \frac{U_o}{r^3} - 3\sigma_{eff} \cdot \frac{1}{r} + P_{neg} \right) + P_{neg} - \frac{2\sigma}{r} \geq 0 \quad (23)$$

or

$$\begin{aligned} \left(\frac{1}{r} \right)^3 - 2\sigma \cdot \frac{4\pi}{3P_{crit} V_o} \cdot \left(1 + \frac{3}{2} \cdot \frac{P_{crit} V_o}{U_o} \cdot \frac{\sigma_{eff}}{\sigma} \right) \left(\frac{1}{r} \right) \\ + P_{neg} \cdot \frac{4\pi}{3P_{crit} V_o} \cdot \left(1 + \frac{P_{crit} V_o}{U_o} \right) \geq 0 \end{aligned} \quad (24)$$

Choosing the equality sign will give an equation whose roots correspond to the intersection(s) of the Fig. 2 curves. This equation is also a reduced cubic, and substitution of the coefficients into the condition $(p/3)^3 + (q/2)^2 = 0$ gives a threshold, or minimum value of P_{neg} , for continued growth of the bubble:

$$\begin{aligned} (2\sigma)^3 \cdot \left(\frac{4\pi}{3P_{crit} V_o} \right)^3 \cdot \left(1 + \frac{3}{2} \frac{P_{crit} V_o}{U_o} \frac{\sigma_{eff}}{\sigma} \right)^3 \cdot \left(\frac{1}{3} \right)^3 \\ = P_{neg}^2 \cdot \left(\frac{4\pi}{3P_{crit} V_o} \right)^2 \cdot \left(1 + \frac{P_{crit} V_o}{U_o} \right)^2 \cdot \left(\frac{1}{2} \right)^2 \end{aligned}$$

or

$$P_{neg} = \sqrt{\frac{128\pi\sigma^3}{81P_{crit} V_o}} \cdot \left[1 + (3/2) \left(P_{crit} V_o / U_o \right) \left(\sigma_{eff} / \sigma \right) \right]^{3/2} \cdot \frac{1}{1 + P_{crit} V_o / U_o} \quad (25)$$

The first term on the RHS is the same as the constant mT solution, Eq. (17b). The second term is the correction for the loss of internal energy, and therefore pressure, to overcome the surface tension and adiabatically form a new surface. The third term is a correction to the second one, representing the contribution of energy by the PV work of the external negative pressure.

At this point, it is in order to confess a hidden approximation in the third term. Setting the PV work equal to $P_{neg} \cdot V(r)$ implies that the integral of PdV has been taken from zero to r . One could make an argument that the integration interval should begin not at zero, but at the radius of a sphere with the

initial volume V_o . Such a correction can be made, but usually turns out to be small (see the discussion of Table 10, below) and, in my opinion it is not worth the added algebraic complication in Eq. (25).

As usual for a reduced cubic with $(p/3)^3 + (q/2)^2 = 0$, there are two equal real roots (i.e., values of $1/r$ for which the equality in Eq. (24) is satisfied. The value of those roots is $\sqrt[3]{2/q}$ (Korn and Korn 1968). Therefore, the value of r at which the bubble is in static equilibrium at the threshold negative pressure is $\sqrt[3]{2/q}$, so

$$r_{eq} = \sqrt[3]{\frac{2}{q}} = \sqrt[3]{\frac{3 P_{crit} V_o}{2 \pi P_{neg} (1 + P_{crit} V_o / U_o)}} .$$

Equation (25) can be manipulated to

$$P_{crit} V_o = \frac{128 \pi \sigma^3}{81 P_{neg}^2} \frac{[1 + (3/2)(\sigma_{eff}/\sigma)(P_{crit} V_o / U_o)]^3}{(1 + P_{crit} V_o / U_o)^2} ,$$

and when this is substituted into the above expression for r_{eq} , the result is

$$r_{eq} = \sqrt[3]{\frac{64}{27} \cdot \frac{\sigma^3}{P_{neg}^3} \cdot \frac{[1 + (3/2)(\sigma_{eff}/\sigma)(P_{crit} V_o / U_o)]^3}{(1 + P_{crit} V_o / U_o)^2}}$$

or

$$r_{eq} = \frac{4\sigma}{3 P_{neg}} \cdot \frac{1 + (3/2)(\sigma_{eff}/\sigma) P_{crit} V_o / U_o}{1 + P_{crit} V_o / U_o} . \quad (26)$$

The first term in this expression is the same as the equilibrium radius in the constant mT cases, Eq. (13). The second term, which is slightly greater than unity, accounts for the lower internal pressure arising from the loss of internal energy during growth that is described by Eq. (22).

To investigate the effect of the $Td\sigma/dT$ term, Eq. (26) was calculated with $\sigma_{eff} = \sigma - Td\sigma/dT$ and with $\sigma_{eff} = \sigma$. The results are shown in Table 8, again omitting cyclohexanol.

When the two sets of calculations, one with and one without the $Td\sigma/dT$ term, are fitted to the measured results, the correlation coefficients are as seen in Table 9.

Table 8. Equation (25) results with and without the $Td\sigma/dT$ term

Material	Recoil energy, keV	Calculated threshold, bars		Measured threshold (bars)
		$Td\sigma/dT$ included	$Td\sigma/dT$ not included	
Isopentane	101	2.316	2.060	3.344
Freon 113	101	2.336	2.115	4.490
Freon 113	166	2.994	2.711	5.207
n-hexane	101	3.547	3.031	6.890
Acetone	101	4.544	4.188	8.309
Trichloroethylene	101	6.031	5.587	11.754
Ethylene bromide	101	9.664	9.197	22.190
Acetophenone	101	12.619	12.322	33.235

**Table 9. Correlation coefficients with
and without the $Td\sigma/dT$ term**

σ_{eff}	R^2
$\sigma - Td\sigma/dT$	0.987
σ	0.992

Both correlation coefficients are high, but the simple σ version (i.e., without the $Td\sigma/dT$ term) is a better straight-line fit. That term is therefore now omitted from Eq. (25), leading to

$$P_{neg} = \sqrt{\frac{128 \pi \sigma^3}{81 P_{crit} V_o}} \cdot \frac{[1 + (3/2)(P_{crit} V_o / U_o)]^{3/2}}{(1 + P_{crit} V_o / U_o)} \quad (27a)$$

$$= \sqrt{\frac{128 \pi \sigma^3 H_{ea}}{81 E_{recoil} P_{crit} / \rho_{crit}}} \cdot \frac{[1 + (3/2)(P_{crit} V_o / U_o)]^{3/2}}{(1 + P_{crit} V_o / U_o)} \quad (27b)$$

The σ_{eff}/σ term should be omitted from Eq. (26) as well. Results calculated from Eq. (27) are plotted in Fig. 6.

A comparison of Tables 7 and 8 shows that while including the effect of varying internal energy may have led to a more self-consistent physical model, it has made little quantitative difference to the predicted thresholds.

Fig. 6. $P_{neg}(\text{exptl})$ vs $P_{neg}[\text{calc, mT model, Eq. (27)}]$. Static terms only. Cyclohexonal excluded. $R^2 = 0.992$.

The reason that the differences between the results from Eq. (17) and from Eq. (27) are so small is surprising, to me at least: only a very small fraction ($\sim 5\%$) of the thermal energy deposited by the recoiling particles is actually used to form new surface area on the expanding bubble.

A simple numerical calculation illustrates this; the algebraic demonstration is tedious and long. The basis of the calculation is Eq. (19), but for the reasons given above, the W_{s2} term is omitted. The integrations of surface energy and PV work are again approximated by beginning the integration at zero radius. Equation (19) becomes

$$U(r) = U_o - W_{s1} - W_e .$$

W_e , the work done in expanding against the external pressure, is actually a negative quantity, work being done by the negative pressure. At the equilibrium radius,

$$U(r_{eq}) = U_o - 4\pi r_{eq}^2 \sigma + \frac{4}{3} \pi r_{eq}^3 P_{neg} , \quad (28)$$

where r_{eq} is given by Eq. (26) and P_{neg} by Eq. (27).

The numerical results of evaluating Eq. (28) are shown in Table 10.

The surface energy typically amounts to only about 10% of the initial internal energy, and almost half of it is provided by the PV work that the negative pressure does on the growing bubble. Therefore, the internal energy at any stage of the growth to the equilibrium radius is little different from that in the simpler, constant mT model.

Table 10. Initial internal energy, surface energy, and PV work

Material	Recoil energy, keV/fJ	U_o , fJ	W_{sl} , fJ	W_e , fJ
Isopentane	101/16.18	15.76	1.8	0.8
Freon 113	166/26.59	25.94	2.8	1.3
Freon 113	101/16.18	15.78	1.7	0.8
n-hexane	101/16.18	15.88	1.3	0.6
Acetone	101/16.18	15.78	1.7	0.8
Trichloroethylene	101/16.18	15.75	1.9	0.8
Ethylene bromide	101/16.18	15.82	1.6	0.7
Acetophenone	101/16.18	15.97	0.9	0.4
Cyclohexanol	101/16.18	15.99	0.9	0.4

Note that W_e , which I approximated as $P_{neg}V$, is small compared with U_o . Therefore, the effect of making the same approximation in the derivation of Eq. (20), and in other equations following from it, must be small.

7.3 DYNAMIC TERMS

Figure 6 showed a strong correlation between the results from Eq. (27) and the measured value of the radiation-induced cavitation threshold when the result from high viscosity cyclohexanol was excluded. However, Fig. 7 shows that when the cyclohexanol result is included, the correlation between measurements and results calculated using only the static bubble formation energy terms is poor. This section gives an approximate treatment, based on simple physical arguments, of the dynamic energy losses.

Riepe and Hahn (1961) estimated the viscous and kinetic energy losses, W_5 and W_4 respectively, by assuming that the nucleation center grew at a velocity not exceeding that required to reach the critical radius r_{crit} within the thermal relaxation time τ_{crit} of a thermal spike of radius r_{crit} . They point out that at the end of that time, the heat content of such a spike would have fallen, through thermal conduction, by a factor $1/e$. The relaxation time is given by $\tau_{crit} = r_{crit}^2/4D$ where D is the thermal diffusivity of the medium.

However, in the mT model of cavitation nucleation, there is no exact equivalent to the critical radius in the Seitz bubble chamber theory; it has been replaced by a criterion that the expansive forces exceed the compressive ones at all radii.

Furthermore, the Riepe and Hahn calculations on bubble chamber nucleation energy agreed with the values deduced from their experimental observations to within 10%. Such close agreement is difficult to reconcile with a situation in which a fraction $1-1/e$, or more than 60%, of the thermal energy might escape the critical radius before the bubble reaches that size.

In fact, the growth could actually take place in a much shorter time than τ_{crit} without upsetting the energy balance significantly because the dynamic terms are relatively small in most cases—see Tables 1 and 4—and less thermal energy would be lost in that shorter time. Also, the energy that does escape by

Fig. 7. $P_{neg}(exptl)$ vs $P_{neg}[calc, mT \text{ model, Eq. (27)}]$. Static terms only. Cyclohexanol included. $R^2 = 0.793$.

thermal conduction may continue to facilitate nucleation by heating the liquid around the expanding bubble, thereby reducing its surface tension and the demands on the bubble's internal energy and pressure: i.e., the surface formation is not entirely adiabatic (in which case the additional $Td\sigma/dT$ surface energy term may indeed not be appropriate).

A slightly different model for the dynamic losses is adopted here. Rather than considering only the time $t(r)$ taken to grow to a particular radius, the growth to any radius r is required to take place in a time $\tau(r)$ or less, that is,

$$t(r) \leq r^2/4D \quad , \quad (29)$$

and therefore, by differentiation the requirement is

$$\dot{r} = \frac{dr}{dt} \geq \frac{2D}{r} \quad , \quad (30)$$

the equality sign being chosen as the limiting case. The viscous loss, which Riepe and Hahn found to be much larger than the kinetic energy loss in most cases, is given in their Eq. (10) as:

$$W_5 = 16\pi\eta \int_0^r \dot{r} r dr ,$$

or, substituting for $\dot{r}$ from Eq. (30),

$$W_5 = 16\pi\eta \int_0^r \frac{2D}{r} r dr = 32\pi\eta Dr . \quad (31)$$

Once again, the integration interval is begun at zero, although the initial radius of the fluid at the critical point could have been used.

The result is different from Riepe and Hahn's Eq. (11), which reads $W_5 = 64\pi\eta Dr$ by a factor of two; the reason is that they assumed a constant growth rate, whereas Eq. (29) allows a lower growth rate in the later stages, when the larger radius would lead to a longer thermal relaxation time.

To calculate the kinetic loss W_4 , the formula used at the critical radius by Riepe and Hahn [Eq. (8) in their paper] is applied here at all radii, with a factor $Mwt/10^3$ to account for the change from mass to molar density units.

$$W_4 = 2\pi\rho_\ell(Mwt/10^3)\dot{r}^2 r^3$$

and substituting for $\dot{r}$ from Eq. (30) gives

$$W_4 = 8\pi\rho_\ell(Mwt/10^3)D^2 r . \quad (32)$$

Again, this differs from Riepe and Hahn's Eq. (11), this time by a factor of four, because the effect of a varying growth velocity has been included.

Note that W_5 and W_4 have the same form and so it is possible, and convenient, to combine them into a single dynamic energy loss term, W_{dyn} , where

$$\begin{aligned} W_{dyn} &= [32\pi\eta D + 8\pi\rho_\ell(Mwt/10^3)D^2] \cdot r \\ &= 32\pi D[\eta + \rho_\ell D(Mwt/10^3)/4] \cdot r \end{aligned}$$

or

$$W_{dyn} = 32\pi\eta_{eff} Dr , \quad (33)$$

where

$$\eta_{eff} = \eta + \rho_\ell DMwt/(4 \cdot 10^3) \quad (34)$$

Equation (33) is the work that would be done against an inwardly directed dynamic pressure P_{dyn} where

$$P_{dyn} = 8\eta_{eff} \frac{D}{r^2} ,$$

because

$$\int_0^r \frac{8\eta_{eff} D}{r^2} \cdot 4\pi r^2 dr = 32\pi\eta_{eff} Dr \quad .$$

In confirmation, by substituting from Eq. (29), Eq. (35) can be rewritten

$$P_{dyn} = 4\eta_{eff} \frac{\dot{r}}{r} \quad (35)$$

which is identical in form to Seitz's expression for the effective pressure at the bubble surface due to viscosity [Seitz 1958, Eq. (39)].

When the dynamic energy term is included, Eq. (22), without the Bugg term, becomes

$$P_{int}(r) = \frac{P_{crit} V_o}{U_o V(r)} \cdot [U_o - 4\pi r^2 \sigma - 32\pi\eta_{eff} Dr + P_{neg} V(r)] \quad , \quad (36)$$

and inclusion of the dynamic pressure term in Eq. (8) gives, for negative external pressures, the following condition for continued growth.

$$P_{int}(r) + P_{neg} \geq \frac{2\sigma}{r} + \frac{8\eta_{eff} D}{r^2} \quad (37)$$

Combining Eqs. (36) and (37) yields

$$\begin{aligned}
 & \left(\frac{1}{r} \right)^3 - 8\eta_{eff} D \cdot \left(1 + 3 \frac{P_{crit} V_o}{U_o} \right) \left(\frac{1}{r} \right)^2 \\
 & - 2\sigma \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{3 P_{crit} V_o}{2 U_o} \right) \left(\frac{1}{r} \right) \\
 & + P_{neg} \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{P_{crit} V_o}{U_o} \right) \geq 0 .
 \end{aligned} \tag{38}$$

Given the assumption that $\sigma_{eff} = \sigma$, the coefficients for $(1/r)^3$ and the constant term are identical to those in Eq. (24). The viscous and kinetic energy effects are contained only in the $(1/r)^2$ term, which was absent from Eq. (24).

Once again, the equality sign in Eq. (38) characterizes the limiting value, or cavitation threshold, of P_{neg} .

Substituting $y=a/3$ for x in the cubic

$$x^3 + ax^2 + bx + c = 0 \tag{39}$$

transforms it into the reduced cubic

$$y^3 + py + q = 0 \tag{40}$$

where

$$p = -a^2/3 + b \tag{41a}$$

and

$$q = 2(a/3)^3 - ab/3 + c . \tag{41b}$$

(See, for example, Korn and Korn 1968).

In this case

$$a = -8\eta_{eff} D \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{3 P_{crit} V_o}{U_o} \right) \tag{42a}$$

$$b = -2\sigma \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{3}{2} \frac{P_{crit} V_o}{U_o} \right) \quad (42b)$$

and

$$c = P_{neg} \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{P_{crit} V_o}{U_o} \right) \quad (42c)$$

The minimum condition for continuous growth is the same as before, viz

$$(p/3)^3 + (q/2)^2 = 0 \quad (43)$$

Given the fluid properties and the energy available for nucleation, which defines U_o through Eq. (18), TK Solver easily calculates P_{neg} from Eqs. (41) and (43), but a little further algebraic manipulation leads to a single equation that can be evaluated with a pocket calculator (or a slide rule).

Start by rearranging Eq. (43):

$$\left(\frac{q}{2} \right)^2 = - \left(\frac{p}{3} \right)^3 = \left(\frac{-p}{3} \right)^3$$

or

$$q = 2(-p/3)^{3/2} \quad .$$

Substitute for q and p from Eq.s (41a) and (41b),

$$\begin{aligned} 2(a/3)^3 - ab/3 + c &= 2 \left(\frac{-a^2/3 + b}{3} \right)^{3/2} \\ &= 2 \left(\frac{-a^2 + 3b}{9} \right)^{3/2} \\ &= \frac{2}{27} (-a^2 + 3b)^{3/2} \quad , \end{aligned}$$

rearranging

$$c = \frac{2}{27}(-a^2 + 3b)^{3/2} - \frac{2a^3}{27} + \frac{ab}{3}$$

Substituting for c from Eq. (42c), and isolating P_{neg} on the left hand side yields

$$P_{neg} = \frac{3P_{crit} V_o}{4\pi(1 + P_{crit} V_o/U_o)} \cdot \frac{1}{27} [2(a^2 - 3b)^{3/2} - 2a^3 + 9ab] , \quad (44)$$

where a, b are defined by Eqs. (42a) and (42b).

As a check of the manipulations, one can set $a = 0$, which means there are no dynamic effects: Eq. (44) then reduces to Eq. (27).

One result from Eq. (44) is surprising: see Table 11. Comparing the calculated thresholds without dynamic effects, Eq. (27), and with dynamic effects, Eq. (44), most of the results show only a small increase, as expected. However, the highly viscous cyclohexanol shows an enormous increase and predicts a threshold almost 50 times greater than the observed value.

There is a simple and plausible physical explanation for this gross overestimation of the cyclohexanol threshold, one which also leads to a different method of calculating the threshold for highly viscous liquids.

Although the mT model of bubble nucleation under negative pressure does not involve a single, critical radius, such as can be identified in the theory of bubble chamber nucleation, there is one "magic" radius that has particular significance for the calculation of dynamic energy losses. Recall that the bubble growth velocity was chosen so that the time taken to grow to a radius r was no greater than the relaxation time of a spherical thermal spike of that radius, thus retaining the thermal energy in the neighborhood of

Table 11. Effect of dynamic terms on the calculated threshold

Material	Recoil energy, keV	Threshold, bars		
		Eq. (27)	Eq. (44)	Measured
Isopentane	101	2.06	2.25	3.344
Freon 113	166	2.12	2.41	4.490
Freon 113	101	2.71	3.21	5.207
n-hexane	101	3.30	3.73	6.890
Acetone	101	4.19	4.69	8.309
Trichloroethylene	101	5.59	6.54	11.754
Ethylene bromide	101	9.20	13.06	22.190
Acetophenone	101	12.32	26.66	33.235
Cyclohexanol	101	9.96	2,779	48.740

the bubble to assist the growth. But, as Eq. (9) showed, once the bubble reaches a radius $r^* = 2\sigma/P_{neg}$ it could continue to grow under the influence of the negative pressure alone, even if no internal energy were left at all. Therefore, a criterion on minimum growth velocity need not be applied to growth beyond a radius of r^* .

When the dynamical terms are small, the equilibrium radius, at which compressive and expansive pressures are equal, is approximately $4\sigma/3P_{neg}$, as given by Eq. (26). In such a case, $r^* = 2\sigma/P_{neg}$ is greater than r_{eq} by a factor of about 1.5, so the growth velocity criterion must be applied all the way up to the equilibrium radius and beyond, and the derivation of Eq. (44) is appropriate in such a case.

However, in a highly viscous liquid the large $1/r^2$ term in Eq. (38) leads to a larger radius being required to reach equilibrium, and indeed the equilibrium radius may be greater than r^* : in such cases, by imposing the minimum growth velocity criterion throughout the process, one is calculating viscous losses in the growth between r^* and r_{eq} , losses that are not necessary because there is no constraint on the time allowed for growth beyond r^* .

The reduced cubic, Eq. (40), has two equal real roots when

$$(p/3)^3 + (q/2)^2 = 0 ,$$

and this case corresponds to the threshold. Those roots are given by

$$y_2 = y_3 = -\sqrt[3]{-\frac{q}{2}} = \sqrt[3]{\frac{q}{2}} , \quad (45)$$

and reversing the transformation $y - a/3 = x$ that was applied between Eqs. (39) and (40) gives

$$x_1 = x_2 = y_2 - \frac{a}{3} = \sqrt[3]{\left(\frac{q}{2}\right)} - \frac{a}{3} .$$

Comparing Eqs. (39) and (38) shows that $x_1 = x_2 = 1/r_{eq}$,

$$r_{eq} = 1 / \left(\sqrt[3]{\left(\frac{q}{2}\right)} - \frac{a}{3} \right) , \quad (46)$$

where q is given by Eq. (41b) and a is given by Eq. (42a).

The "magic" radius, above which the bubble can grow without further reliance on internal pressure, is given by

$$r^* = 2\sigma/P_{neg} . \quad (47)$$

The two radii corresponding to the cases listed in Table 11 are shown in Table 12.

Table 12. Calculated thresholds, equilibrium radius, and unconditional growth radii

Material	Recoil energy, keV	Threshold, bars, Eq. (44)	r_{eq} , nm Eq. (46)	r^* , nm Eq. (47)
Isopentane	101	2.25	92.8	133.4
Freon 113	166	2.41	103.9	147.1
Freon 113	101	3.21	79.1	110.5
n-hexane	101	3.73	69.8	99.5
Acetone	101	4.69	70.8	100.9
Trichloroethylene	101	6.54	64.2	89.9
Ethylene bromide	101	13.06	45.6	59.4
Acetophenone	101	26.66	27.2	29.6
Cyclohexanol	101	2,779	4.4	0.24

Except for cyclohexanol, r^* is greater than r_{eq} : thus Eq. (44) is appropriate, the rapid growth criterion being applicable up to and beyond r_{eq} . Note also that for the low viscosity, low threshold liquids, the value of r^* is roughly 1.5 r_{eq} as the simple mT model, with static terms only, predicts.

For cyclohexanol, however, the model has imposed a rapid growth requirement in the size range 0.24 nm to 4.4 nm, when in fact that requirement existed only during the growth up to 0.24 nm. One is, of course, entitled to wonder if the cyclohexanol bubble really knows the opportunity for a more leisurely growth that is open to it, but the fact remains that beyond r^* a slow growth, which the viscosity will tend to promote, would not prevent the bubble from exceeding r_{eq} and growing indefinitely.

And, that is not the only problem with applying the basic mT model to very high threshold liquids: Table 13 compares the equilibrium radius r_{eq} and the magic radius r^* with R_o , the radius that the fluid volume V_o would have, at the critical point, if it were spherical.

$$\frac{4\pi R_o^3}{3} = V_o = \frac{M_o}{\rho_{crit}}$$

$$\therefore R_o = \sqrt[3]{\frac{3 V_o}{4\pi}} = \sqrt[3]{\frac{3 E_{recoil}}{4\pi \rho_{crit} H_{ca}}} \quad (48)$$

The implicit assumption that the volume and surface area of the initial nucleation center are small compared with the values during the important stage of growth when $r \sim r_{eq}$ or r^* is unrealistic for acetophenone and completely incorrect for cyclohexanol. Furthermore, for cyclohexanol the calculated equilibrium radius and the calculated magic radius are both smaller than the initial radius of the heated fluid, which makes no sense.

Table 13. Comparison of r_{eq} , r^* , and R_o

Material	Recoil energy, keV	Threshold, bars, Eq. (44)	r_{eq} , nm Eq. (46)	r^* , nm Eq. (47)	R_o , nm Eq. (48)
Isopentane	101	2.25	92.8	133.4	31.0
Freon 113	166	2.41	103.9	147.1	35.8
Freon 113	101	3.21	79.1	110.5	30.3
n-hexane	101	3.73	69.8	99.5	28.9
Acetone	101	4.69	70.8	100.9	27.3
Trichloroethylene	101	6.54	64.2	89.9	27.6
Ethylene bromide	101	13.06	45.6	59.4	25.1
Acetophenone	101	26.66	27.2	29.6	23.5
Cyclohexanol	101	2,779	4.4	0.24	23.2

Although Hahn reported measuring the a-recoil induced cavitation threshold of 28 different compounds, he published the results from only eight of them, as far as I know. There is only one good high viscosity data point to work with, and that one, cyclohexanol, needs to be treated carefully because the measurement is reported as being carried out at a temperature that is below the freezing point. Therefore, I adopted only a very simple, although physically reasonable, approach to calculating the cavitation threshold in such a case, because a large mathematical effort did not seem appropriate for a single data point.

Given the energy available, one can calculate the size (e.g., volume or radius) of a sphere of liquid that can be raised to the critical point, thus providing a region in which homogeneous nucleation and subsequent evaporation will take place. If the negative pressure is high enough that the magic radius, $2\sigma/P_{neg}$, is equal to or less than this initial bubble size, then the initial bubble can grow even if the high viscosity causes a low growth rate and therefore loses heat through conduction. A threshold, P_{neg} , can be calculated by imposing this inequality:

$$\frac{2\sigma}{P_{neg}} \leq R_o$$

In the limiting case or threshold, when the equality holds,

$$P_{neg} = \frac{2\sigma}{R_o} \quad (49)$$

and substituting for R_o from Eq. (48) yields

$$P_{neg} = 2\sigma \sqrt[3]{\frac{4\pi\rho_{crit}H_{ca}}{3E_{recoil}}} \quad (50)$$

This equation can be applied legitimately only to a liquid with high η_{eff} ; a nucleation center in other liquids can grow quickly enough to reach its equilibrium radius, before the initial heat has been dissipated, without paying too high a price in dynamic losses, leading to a lower threshold than indicated by Eq. (50).

How is one to decide which liquids are "high viscosity" and which are not? By calculating the thresholds from both Eq. (44) and Eq. (50): if the threshold from Eq. (44) is lower, then that is the appropriate model to use (i.e., the liquid is classed as low viscosity). Otherwise, Eq. (50) is applied. The results are shown in Table 14.

Figure 8 plots the calculated and measured threshold, including the cyclohexanol results, and a linear fit to the data. Comparing Fig. 8 and Fig. 7 shows that the inclusion of dynamic effects has greatly improved the fit of the complete data set (i.e., including cyclohexanol). The correlation coefficient is not as high as for the simpler, static terms only, equations without cyclohexanol, but the slope is much improved; a perfect fit would have a slope of 1, and the slope of Fig. 6 is 2.81, whereas in Fig. 8 it is 1.46.

Table 14. Threshold calculated from Eqs. (44) and (50)

Material	E_{recoil} keV	Threshold, bars			
		Eq. (44)	Eq. (50)	MIN [Eq. (44), Eq. (50)]	Measured
Isopentane	101	2.25	9.68	2.25	3.34
Freon 113	166	2.41	9.90	2.41	4.49
Freon 113	101	3.21	11.68	3.21	5.21
n-hexane	101	3.73	12.83	3.73	6.89
Acetone	101	4.69	17.36	4.69	8.31
Trichloroethylene	101	6.54	21.31	6.54	11.75
Ethylene bromide	101	13.06	30.92	13.06	22.19
Acetophenone	101	26.66	33.65	26.66	33.23
Cyclohexanol	101	2,779	28.98	28.98	48.74

Fig. 8. P_{neg} (exptl) vs P_{neg} [calc, variable mT model, Eqs. (44) and (50)]. Static and dynamic terms. Cyclohexonal included. $R^2 = 0.960$.

8. AVAILABLE ENERGY

So far, I have assumed that all of the recoil energy is available for bubble nucleation. However, there are some hints in the results presented earlier that this may not be so.

8.1 ENERGY DISCREPANCIES

The first observation is that the measured thresholds in Freon 113 with two very different recoil energies (166 keV and 101 keV) differ by a factor of only 1.16 (according to my measurements, which were necessarily not very precise, from Hahn's published graphs), whereas the values calculated from Eq. (44) differ by a factor 1.33. This would be consistent with several possible explanations, including one that involves less than the full energy of the more energetic recoil being available to a single nucleation center.

The second observation is that all of the calculated values in, for example, Table 14 are lower than the corresponding measured values. This, too, is consistent with a situation in which less than the full recoil energy is actually available. But it is consistent with several other possibilities too.

8.2 LINEAR RATE OF ENERGY LOSS

Riepe and Hahn calculated the available energy by considering the energy deposited by the recoiling nucleus and the alpha particle within a length equal to the critical bubble diameter. In the mT -model, there is no single, critical bubble size, and so a different approach is needed.

The linear rate of energy loss, dE/dx , of the heavy, slow, recoiling nucleus is one to two orders of magnitude higher than that of the ejected alpha particle, and in the following calculations, any contribution to the nucleation energy from the alpha is neglected.

The heavy (mass > 200 amu) recoils from natural α -decay all have initial velocities less than electron orbital velocities, and a simple, although approximate, expression for their rate of energy loss is the Bohr formula

$$\frac{dE}{dx} = \frac{\pi \hbar^2}{\mu} Z_p^{2/3} Z_\ell N \rho_\ell, \quad (51)$$

where μ is the electron mass (9.109×10^{-31} kg); $\hbar$ is the Planck constant divided by 2π (1.055×10^{-34} J), Z_p is the charge number of the recoil nuclei (82 for ^{210}Po and ^{212}Po decays), Z_ℓ is the charge number of the liquid molecules, N is Avogadro's number (6.022×10^{23} mol $^{-1}$), and ρ_ℓ is the molar density of the liquid.

This version of the formula was taken, with a change of symbols and units, from Bohr's book (Bohr 1948); the same formula was used by Riepe and Hahn (1961). Note that according to Eq. (51), in this regime the rate of energy loss, as calculated from the Bohr formula, is independent of energy.

Substituting the physical constants into Eq. (51) yields

$$\frac{dE}{dx} = 4.36 \times 10^{-13} \rho_\ell Z_\ell. \quad (52)$$

The units of dE/dx will be joules/metre, if ρ_ℓ is expressed in mols/m 3 .

Remember, this formula applies only to the recoils from the α -decay of Po: a different constant would apply to the nuclei of other elements, because Z_p would be different.

Apfel and his colleagues (Apfel, Sun, and Ravinda 1992) have made much more detailed studies of energy deposition, with computer simulations of both thermal and mechanical effects, than the simple approach of this report.

8.3 NUCLEATION CENTER FORMATION

As a simplified model, consider that this linear rate of energy deposition could create a cylinder of fluid at the critical temperature and pressure. In the nomenclature of the previous sections, if d is the average diameter of the heated cylinder, then

$$\frac{\pi}{4} d^2 \rho_{crit} H_{ca} = \frac{dE}{dx} = 4.36 \times 10^{-13} \rho_\ell Z_\ell$$

or

$$d = 2 \sqrt{\frac{dE/dx}{\pi \rho_{crit} H_{ca}}} \quad (53)$$

First, suppose that the recoil particle has a range that is long compared with the diameter d ; actually, in many cases it is not so long, but that situation is easy to deal with. Then, imagine that the long cylinder of heated vapor breaks up into separate bubbles, perhaps because of the severe irregularities in its actual diameter (because the recoil loses its energy in a relatively small number of statistically distributed collisions, the energy deposition in any short segment of the track is likely to differ significantly from the average value and, of course, the axis of the cylinder will not be straight).

If the length of the cylinder deforming into a spherical bubble is short (Fig. 9a), then the diameter of the spherical bubble is greater than that length, and some of the small bubbles will touch and coalesce into larger ones.

If, on the other hand, the length of the cylindrical section is much greater than its diameter (Fig. 9b), the resulting sphere has a diameter smaller than the length of its cylindrical progenitor, and in that case the adjacent spheres will not touch and coalesce.

The limiting case, with the spherical bubblets arrayed like pearls on a string, just touching each other, occurs when

$$\frac{\pi d^2}{4} \ell = \frac{4\pi}{3} \left(\frac{\ell}{2}\right)^3 \quad (54)$$

or

$$\ell = \sqrt{\frac{3}{2}} d \approx 1.225 d \quad (55)$$

Cylinder Length

a. 0.33 X diameter

b. 3 X diameter

c. 1.225 X diameter

Fig. 9. Unit diameter cylinders and spheres of the same volume.

See Fig. (9c). Or, substituting for d from Eq. (53),

$$\ell = \sqrt{\frac{6 dE/dx}{\pi \rho_{crit} H_{ca}}} \quad (56)$$

The energy available to form this bubble is $\ell dE/dx$, the length of track within the cylindrical section $\times$ the linear energy deposition rate, so

$$E_{av} = \ell \frac{dE}{dx} = \sqrt{\frac{6 (dE/dx)^3}{\pi \rho_{crit} H_{ca}}} \quad (57)$$

The volume of the cylindrical section, and of the spherical bubble formed from it, is

$$V_o = \pi \frac{d^2}{4} \ell = \pi \sqrt{\frac{3}{32}} d^3 \quad ,$$

or, substituting for d from Eq. (53),

$$V_o = \sqrt{\frac{6}{\pi}} \left(\frac{dE/dx}{\rho_{crit} H_{ca}} \right)^3 \quad (58)$$

Bubbles formed from cylindrical sections shorter than the length defined by Eq. (55) will touch and coalesce, thereby growing. But what about longer sections—could a line of larger, but separated, bubbles form?

My argument is that the irregularities in the recoil particle's track will create localized regions of higher and lower energy deposition, on a scale small compared with the total track length. These localized regions will form tiny overlapping bubbles, each smaller than the size given by Eq. (56).

Adjacent bubbles will coalesce, but once a bubble of diameter $\sqrt{3/2}d$ has been formed in this way, it will not be in contact with the adjacent bubble of the same size or smaller. Therefore, there will be no more coalescence. Equation (56) therefore represents the volume of the largest nucleation centers along the particle's track.

The mass of fluid in the bubble is $M_o = V_o \rho_{crit}$

$$\therefore M_o = \sqrt{\frac{6}{\pi}} \left(\frac{dE/dx}{H_{ca}} \right)^3 \quad (59)$$

For good physical reasons, exactly the same algebraic results follow from asking the simpler question, "What size of sphere can be raised to the critical point by the energy deposited along a length equal to its own diameter?" In this case, $M_o H_{ca} = \text{diameter} \times dE/dx$, or

$$V_o \rho_{crit} H_{ca} = \sqrt[3]{\frac{6V_o}{\pi}} \cdot dE/dx ;$$

therefore,

$$V_o = \sqrt{\frac{6}{\pi} \left(\frac{dE/dx}{\rho_{crit} H_{ca}} \right)^3} ,$$

which is the same as Eq. (58).

The energy that might be used in place of the full recoil energy, in the equations derived earlier in this report, is $\ell dE/dx$, given by Eq. (57). However, in some cases the actual track length of the recoil, according to Bohr's formula, is less than the length ℓ . In that event, the quantity $\ell dE/dx$ would be greater than E_{recoil} , but of course only the recoil energy is actually available. In the language of TK Solver, this can be written

$$E_{av} = MIN(\ell dE/dx, E_{recoil}) , \quad (60)$$

or, in words, the available energy is the lesser of $\ell dE/dx$ and E_{recoil} . Substituting for $\ell dE/dx$ from Eq. (57) yields

$$E_{av} = MIN \left(\sqrt{\frac{6(dE/dx)^3}{\pi \rho_{crit} H_{ca}}} , E_{recoil} \right) . \quad (61)$$

To show the impact of these considerations, Table 15 compares the recoil energy with $\ell dE/dx$, and E_{av} , calculated as above, for the liquids whose cavitation threshold was reported by Hahn.

Table 15. Energy available for bubble formation

Material	Energy, keV		
	E_{recoil}	$\ell dE/dx$ Eq. (54)	E_{av} Eq. (61)
Isopentane	101	47	47
Freon 113	166	139	139
Freon 113	101	139	101
n-hexane	101	47	47
Acetone	101	52	52
Trichloroethylene	101	110	101
Ethylene bromide	101	158	101
Acetophenone	101	58	58
Cyclohexanol	101	56	56

8.4 CALCULATION OF CAVITATION THRESHOLD

Write

$$\frac{dE}{dx} = \frac{\pi \hbar^2}{\mu} Z_p^{2/3} Z_\ell N \rho_\ell \quad \text{from Eq. (51),}$$

$$V_o = \sqrt{\frac{6}{\pi} \cdot \left(\frac{dE/dx}{\rho_{crit} H_{ca}} \right)^3} \quad \text{from Eq. (58),}$$

$$E_{av} = MIN \left(\sqrt{\frac{6 (dE/dx)^3}{\pi \rho_{crit} H_{ca}}}, E_{recoil} \right) \quad \text{from Eq. (61),}$$

$$U_o = E_{av} - \rho_{crit} V_o \quad \text{from Eq. (18),}$$

$$a = -8\eta_{eff} D \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + 3 \frac{P_{crit} V_o}{U_o} \right) \quad \text{from Eq. (42a),}$$

$$b = -2\sigma \cdot \frac{4\pi}{3 P_{crit} V_o} \cdot \left(1 + \frac{3}{2} \frac{P_{crit} V_o}{U_o} \right) \quad \text{from Eq. (42b).}$$

Then the calculated thresholds for low and high viscosity situations are derived by substituting the above expressions into Eqs. (44) and (50). From Eq. (44),

$$P_{neg}(62) = \frac{3 P_{crit} V_o}{4\pi (1 + P_{crit} V_o / U_o)} \cdot \frac{1}{27} [2(a^2 - 3b)^{3/2} - 2a^3 + 9ab] , \quad (62)$$

and from Eq. 50, substituting E_{av} for E_{recoil} ,

$$P_{neg}(63) = 2\sigma \sqrt[3]{\frac{4\pi P_{crit} H_{ca}}{3 E_{av}}} . \quad (63)$$

The calculated negative pressure threshold for radiation-induced cavitation is

$$P_{neg}(threshold) = MIN[P_{neg}(62) , P_{neg}(63)] . \quad (64)$$

Table 16 shows the results from these equations, including E_{av} , $P_{neg}(62)$, and $P_{neg}(63)$.

The calculated thresholds [Eq. (64)] and the experimental values are plotted in Fig. 10: there is a reasonable straight line fit, with a slope of almost exactly unity and only a small intercept.

The details of the fit are not as good as some of the earlier, simpler equations—in particular, the calculated values for isopentane and Freon 113 are badly out of order—but the high viscosity cyclohexanol has been successfully brought into the fit. And the average slope is now right.

It is perhaps worth remarking that the Bohr formula for the dE/dx of slow, heavy particles is well recognized to be only an approximation. Riepe and Hahn quoted results from another formula differing from the Bohr approximation by as much as 15% and also experimental results that differed from the calculations by more than 50%. Such a difference, for example, would change the calculated values of some thresholds by as much as 30%.

Furthermore, it is an assumption of Bohr's range calculation that the mass of the energetic recoil particle is very much greater than the mass of the nuclei in the medium it is traversing; otherwise, the particle would essentially diffuse rather than pursue a definite track. This assumption is probably valid

Table 16. Calculated and measured cavitation thresholds

Material	Available energy (keV)	Negative pressure thresholds, bars			Exptl.
		P_{neg} (62)	P_{neg} (63)	P_{neg} (64)	
Isopentane	47	3.42	12.46	3.42	3.34
Freon 113	139	2.66	10.49	2.66	4.49
Freon 113	101	3.21	11.68	3.21	5.21
n-hexane	47	5.81	16.60	5.81	6.89
Acetone	52	6.87	21.69	6.87	8.31
Trichloroethylene	101	6.54	21.31	6.54	11.75
Ethylene bromide	101	13.06	30.92	13.06	22.19
Acetophenone	58	45.51	40.44	40.44	33.23
Cyclohexanol	56	8925.87	35.35	35.35	48.74

Fig. 10. P_{neg} (exptl) vs P_{neg} [calc, variable mT model, Eq. (64), using E_{av}]. Static and dynamic terms. Cyclohexanol included. $R^2 = 0.960$, slope = 1.01, intercept = 2.9.

for the liquids containing only hydrogen, carbon, and oxygen. It is not really valid for liquids containing chlorine or bromine atoms. Therefore, one might expect my calculation of E_{av} to be even more suspect for Freon, trichloroethylene, and ethylene bromide than for the other liquids. It is possible that this observation is relevant to the fact that Eq. (64) wrongly orders the thresholds for isopentane and Freon 113, while also significantly underestimating the thresholds for trichloroethylene and ethylene bromide.

Considering the number of effects to be considered (including homogeneous nucleation thresholds, bubble growth thermodynamics, dynamic effects, and the energy deposition rates of nuclear particles), I regard Fig. 10 as a good result, especially in comparison with the earlier model that produced Fig. 4, which excluded the high viscosity cyclohexanol case and still had a slope of 14 with a correlation coefficient of only 0.66.

For interest, Table 17 shows the continuing improvement in the agreement between calculated and experimental results, in terms of the slope of a graph of one against the other, as the successively more comprehensive models in this report are applied. It is, however, interesting, and somewhat puzzling, that the correlation coefficient does not continuously improve, although it probably remains acceptable.

The fact that the intercept of the graphs is not zero is less troublesome. At low thresholds ($P_{neg}/2 < P_{vap}$), the models would not be applicable anyway. Certainly the final model's incorrect predictions that the Freon threshold would be lower than the isopentane one and that the threshold for trichloroethylene would be lower than for acetone, is more disturbing than the nonzero intercept or the lower correlation coefficient.

Table 17. Evolution of the model

Model	Equation	Figure	Cyclohexanol included?	Dynamic effects?	dE/dx effects?	Straight-line fit to experiment		
						Slope	Intercept	R ²
R_{crit}		4	No	No	No	13.9	-13.9	0.66
mT	(17)	5	No	No	No	2.9	-2.5	0.99
mT	(27)	6	No	No	No	2.8	-2.6	0.99
mT	(44), (50)	8	Yes	Yes	No	1.5	1.2	0.96
mT	(64)	10	Yes	Yes	Yes	1.0	2.8	0.86

9. SUMMARY

In 1961, Georg Riepe's Ph.D. research at the University of Fribourg had shown that nucleation of bubbles in the superheated liquid of a bubble chamber was quantitatively explained by the Sietz thermal spike theory—at least for the case of bubble formation by the heavy, slow nuclei recoiling from alpha decay (Riepe and Hahn 1961).

Two years later, another paper from the same laboratory reported that in cold liquids, "superheated" by application of a tensile stress, there was an order of magnitude discrepancy between the experimental results and the predictions of the same theory that had served Riepe so well (Hahn and Peacock 1963).

In 1967, a new report, from a different laboratory, provided a physical model of the criteria for bubble formation that offered a reason for the difference in behavior under irradiation of hot superheated liquids and cold ones, but the quantitative predictions for bubble nucleation under negative pressures, although improved over the earlier theory, were still in poor agreement with the experimental data (West 1967).

The present report amends and extends the 1967 theory, here called the mT model. Predictions and experimental data, from the few measurements available of bubble formation by slow, heavy particles in liquids under negative pressure, are now in reasonably good agreement, even for those liquids where dynamic effects (viscous and kinetic energy losses) are significant or dominant. For this author, it provides some satisfaction to see progress in a puzzle that has been bothersome for more than 30 years.

The initial impetus for the work described here arose from the Spallation Neutron Source (SNS) Project. The SNS will have a liquid mercury target which may, under certain circumstances, be subject to negative pressure transients. I plan a separate, shorter report applying the above theory specifically to mercury.

10. ACKNOWLEDGMENTS

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Many colleagues at Oak Ridge National Laboratory have been kind enough to listen to descriptions of this work and to offer helpful criticism, advice, and ideas. I thank them for their help and interest. D. M. Williams kindly prepared the drawings in Fig. 9. L. K. Mansur recognized, and pointed out to me, the analogies between the original mT model and a theory later developed to describe onset of swelling in irradiated solids and its dependence upon the helium created in neutron-induced transmutation reactions.

Because of the potential significance of radiation-induced cavitation in the liquid mercury target of the proposed new SNS, John Haines of that project's target team kindly provided encouragement for the work and support for the publication of this report.

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12. LIST OF SYMBOLS

Symbol	Definition
α	Ratio of the vapor pressure over a concave surface of radius r_{crit} to that over a plane surface
a	Coefficient of the quadratic term in a nonreduced cubic: Eq. (39)
b	Coefficient of the linear term in a nonreduced cubic: Eq. (39)
c	Constant in a nonreduced cubic: Eq. (39)
C_ℓ	Specific heat of the liquid
d	Average diameter of heated cylinder
σ	Surface tension
σ_{eff}	Effective surface tension including the Bugg term
D	Thermal diffusivity
dE/dx	Linear rate of energy deposition
E_{av}	Energy available to form the nucleation center
E_{recoil}	Energy of recoil from α -decay
$\hbar$	Planck's constant $\div 2\pi$
H_{ca}	Enthalpy difference between ambient conditions and the critical point
$H(T)$	Enthalpy at temperature T
H_v	Enthalpy of evaporation
H_{vr}	Enthalpy of evaporation from a concave surface of radius r
$H_{v\infty}$	Enthalpy of evaporation from a plane surface
ℓ	Length of heated cylinder
M_o	Quantity (mols) of liquid raised to the critical point
$M(r)$	Quantity (mols) of vapor inside the bubble when its radius is r
Mwt	Molecular weight
N	Avogadro's number
η	Liquid viscosity
η_{eff}	Effective viscosity including kinetic energy losses
p	Coefficient of linear term in a reduced cubic
ρ_{crit}	Fluid density at the critical point
ρ_ℓ	Liquid density
ρ_v	Vapor density
P_{crit}	Critical pressure
$P_{int}(r)$	Pressure inside the bubble when its radius is r
P_{ext}	Liquid pressure outside the bubble
P_{neg}	Magnitude of the negative pressure in the liquid outside the bubble
P_{vap}	Vapor pressure over a plane surface
P_{vr}	Vapor pressure over a concave surface of radius r
P_{vrcrit}	Vapor pressure over a concave surface of radius r_{crit}
q	Constant in a reduced cubic
r	Bubble radius
$\dot{r}$	dr/dt
r^*	The "magic" radius, $2\sigma/P_{neg}$
r_{crit}	Critical radius for continued growth: Eq. (1)
r_{eq}	In the mT model, the bubble radius at which compressive and expansive terms are equal
R	Molar gas constant
R_o	Radius of a spherical mass M_o at the critical point
$R^2, R^{\wedge 2}$	Correlation coefficient of the linear regression

R_r	Recoil range
τ	Relaxation time of a spherical thermal spike
τ_{crit}	Relaxation time of a spherical thermal spike of radius r_{crit}
t	Time
T_{op}	Operating temperature
T_{crit}	Critical temperature
μ	Electron mass
U_o	Initial internal energy
$U(r)$	Internal energy as a function of radius
v_c	Velocity of the bubble wall as it grows to the critical radius
V_o	Volume of the mass M_o at the critical pressure and temperature
$V(r)$	Volume of bubble when its radius is r
W_{s1}	Surface energy
W_{s2}	Bugg surface energy term
W_e	Work done by the expanding bubble on the surrounding liquid
W_v	Enthalpy of evaporation of the vapor filling the bubble
W_4	Kinetic energy given to the liquid moving away as the bubble expands
W_5	Viscous losses in moving away the liquid as the bubble expands
λ	Thermal conductivity of the liquid
y_2, y_3	Real roots of a cubic: Eq. (45)
Zl	Charge number of the liquid molecules
Zp	Charge number of the recoiling nucleus

APPENDIX A

MEASUREMENTS OF THE PRESSURE THRESHOLD FOR BUBBLE NUCLEATION BY VARIOUS PARTICLES IN VARIOUS LIQUIDS

Table A.1. Boiling results

Method	Liquid	Temp. (°C)	Source of energetic particles	Max. energy (α , n, e ⁻ , etc.) (MeV)	Heaviest recoil ^a and max. energy (keV)	Threshold ^b (bars)	Reference
Bubble chamber	Freon 12	29.4	²¹² Po- α	8.784	²⁰⁸ Pb, 166	1.70	Fig. 8, Riepe and Hahn 1961
"	"	"	²¹² Bi- α	6.090	²⁰⁸ Tl, 115	1.95	"
"	"	"	²¹⁰ Po- α	5.304	²⁰⁶ Pb, 101	2.00	"
"	"	40	"	"	"	1.79	Fig. 9, Riepe and Hahn 1961
"	"	50.2	"	"	"	1.55	"
"	"	54.3	"	"	"	1.38	"
"	"	69.2	"	"	"	1.17	"
"	"	79.3	"	"	"	0.88	"
"	Propane	18.5	²¹² Po- α	8.784	²⁰⁸ Pb, 166	1.66	Fig. 12, Riepe and Hahn 1961
"	"	"	²¹² Bi- α	6.090	²⁰⁸ Tl, 115	1.85	"
"	"	"	²¹⁰ Po- α	5.304	²⁰⁶ Pb, 101	1.97	"
"	Freon 13B1	36.1	⁸⁷ Ar	0.0023	(Ba, ϵ ~10 ⁵) ^c	6.22	p. 891, Riepe and Hahn 1961
"	Water	193-227	U(n, fission)	200		1.8 ^d -2.4 ^d	Fig. 6, Dietrich and Connolly 1973
Superheat vessel ^e	Acetone	106 $\pm$ 5	D(d,n) ³ He	2.45	¹⁶ O, 543	3.85-4.93	Tab. 3, El Nagdy and Harris 1973
"	"	91 $\pm$ 5	T(d,n) ⁴ He	14.1	¹⁶ O, 3122	2.57-3.38	"
"	Benzene	140 $\pm$ 5	D(d,n) ³ He	2.45	¹² C, 696	4.22-5.23	"
"	"	120 $\pm$ 5	T(d,n) ⁴ He	14.1	¹² C, 4005	2.64-3.36	"
"	Water	175 $\pm$ 5	"	"	¹⁶ O, 3122	7.9-10.0	"
S.D.D. ^f	Freon 12	34.3	D(d,n) ³ He	4.14	³⁷ Cl, 424	0.82	Apfel 1979
"	Freon 114	34.3	"	"	³⁷ Cl, 424	1.10	"
"	C ₄ H ₈	34.2	"	"	¹² C, 1180	1.5	"
"	Isobutane	20-23°C	T(p,n) ³ He	1.2	¹² C, 341	2.04-2.32	p. 5264, Lo and Apfel 1988
"	Freon 142B	"	"	2.1	³⁷ Cl, 424	1.88-2.17	"
"	Freon 114	"	D(d,n) ³ He	6.0	³⁷ Cl, 615	0.81-1.00	"

^aUsually the decayed nucleus recoiling from the α -emission, or the heaviest knock-on atom in the liquid in the case of neutron irradiation (the heaviest knock-on probably has the highest energy deposition per unit track length [e.g., Apfel, Roy, and Lo (1985), Figs. 1 and 2] and may therefore be the most effective at nucleating bubbles.)

^bFor bubble chamber results, and the Apfel results, this is the difference between the vapor pressure and the external pressure after expansion [i.e., P_{ext} in the language of Eq. (1)]. For the Lo and Apfel superheated drop case, it is the difference between the vapor pressure at 21-23°C and atmospheric pressure. For the superheated drop detector, I recalculated the figures on the basis of more recent data on vapor pressures given by Yaws (1996).

^cTypical fission fragments.

^dThese results were reported as the superheat, in °F, at which bubbles were or were not observed to be nucleated by fission fragments. I estimated the boundary, or threshold, by eye, and took the difference in the vapor pressure (from the Yaws 1996 database) of water between the initial temperature, and the initial temperature plus the threshold superheat.

^eThese results were reported as the superheat, in °C, when the liquid was depressurized to atmospheric pressure. I used the Yaws 1996 vapor pressure data to calculate $P_{vap} - P_{ext}$ from the superheat.

^fCis-2-butene.

Table A.2. Negative pressure results

Liquid	Method	Temp. (°C)	Freq. (kHz)	Source of energetic particles	Negative pressure threshold (bars)	Reference
Freon 113	Spinner	20	n.a.	$^{210}\text{Po}-\alpha$	5.21	Hahn 1961
"	"	"	"	$\text{Th}(\text{C}+\text{C}')-\alpha$	4.49	"
"	"	"	"	$^{137}\text{Cs}-\gamma$	53	"
"	"	"	"	^{252}Th fission	5	Spadavecchia and Hahn 1967
"	"	"	"	$\text{Pu}/\text{Be}-n$	3.0	"
"	"	"	"	"	<5	Hahn 1961
"	Ultrasonic	0	39	"	3.8	Greenspan and Tscheig 1967
"	"	26	35	"	1.8	"
"	"	28	35	"	2.0	"
"	"	29	35	"	1.6	"
"	"	"	45	"	1.8	"
"	"	"	44-52	$\text{D}(\text{d},n)^3\text{He}$	4.4	"
"	"	"	"	$\text{T}(\text{d},\alpha)n$	0.7	"
Methyl alcohol	Ultrasonic	29	44-52	$\text{D}(\text{d},n)^3\text{He}$	10.4	Greenspan and Tscheig 1967
"	"	"	"	$\text{T}(\text{d},\alpha)n$	2.4	"
"	"	27	39	$\text{Pu}/\text{Be}-n$	5.0	"
"	"	"	52	"	5.1	"
"	"	26	37	$\text{ThC}'-\alpha$	11.0	"
"	"	?	?	$\text{Th}/\text{fast fission}$	-10db ^a	"
"	"	?	?	$\text{U}/\text{fission}$	?	"
Ethyl alcohol	Spinner	20	n.a.	^{232}Th fission	0.55	Spadavecchia and Hahn 1967
"	"	"	"	^{238}U fission	0.43	"
"	Ultrasonic	29	44-52	$\text{Pu}/\text{Be}-n$	6.7	Greenspan and Tscheig 1967
"	"	"	"	$\text{D}(\text{d},n)^3\text{He}$	13.4	"
"	"	"	"	$\text{T}(\text{d},\alpha)n$	4.0	"
Freon 11	Spinner	20	n.a.	$^{137}\text{Cs}-\gamma$	≈57	Hahn 1961
"	Ultrasonic	20	25	$\text{Pu}/\text{Be}-n$	<5	Hahn and Peacock 1963
"	"	26.5	44	"	1.7	Greenspan and Tscheig 1967
"	"	27	45	"	1.9	"

Table A.2 (continued)

Liquid	Method	Temp. (°C)	Freq. (kHz)	Source of energetic particles	Negative pressure threshold (bars)	Reference
Methylene chloride	Ultrasonic	26.3	35	Pu/Be-n	3.0	Greenspan and Tscheig 1967
"	"	27.2	43	"	5.1	"
"	"	27.4	29	"	3.3	"
"	"	28.7	53	"	4.5	"
Diethylether	Ultrasonic	24.8	55	Pu/Be-n	1.9	Greenspan and Tscheig 1967
"	"	27.4	54	"	1.4	"
"	"	28	45	Th- α	4.0	"
n-hexane	Spinner	20	n.a.	$^{210}\text{Po-}\alpha$	6.89	Hahn 1961
"	Ultrasonic	26.5	41	Pu/Be-n	2.4	Greenspan and Tscheig 1967
"	"	26.9	35	"	2.5	"
Tetrachloroethylene	Spinner	≈ 20	n.a.	Po/Be-n	6.6 ± 0.25	West and Howlett 1969
"	Ultrasonic	≈ 20	20.5	"	6.5 ± 1.2	"
"	"	27	49	Pu/Be-n	7.5	Greenspan and Tscheig 1967
Acetone	Spinner	20	n.a.	$^{210}\text{Po-}\alpha$	8.31	Hahn 1961
"	Ultrasonic	$\approx 22?$	?	Po/Be-n	5.5	Lieberman 1959
"	"	$\approx 22?$	?	Pu/Be-n	4.8	Greenspan and Tscheig 1967
n-hexane	Spinner	20	n.a.	$^{210}\text{Po-}\alpha$	6.89	Hahn 1961
"	Ultrasonic	26.5	41	Pu/Be-n	2.4	Greenspan and Tscheig 1967
"	"	26.9	35	"	2.5	"
Isopentane	Spinner	20	n.a.	$^{210}\text{Po-}\alpha$	3.34	Hahn 1961
Isopentane?	Ultrasonic	$\sim 22?$	?	Po/Be-n	2.7	Lieberman 1959
"	"	$\sim 22?$	?	e ⁺	>22	"
n-pentane	Ultrasonic	24.5	28	Pu/Be-n	1.5	Greenspan and Tscheig 1967
"	"	26.5	35	"	1.0	"
Trichloroethylene	Spinner	20	n.a.	$^{210}\text{Po-}\alpha$	8.31	Hahn 1961
"	Ultrasonic	30	48	Pu/Be-n	4.5	Greenspan and Tscheig 1967

Table A.2 (continued)

Liquid	Method	Temp. (°C)	Freq. (kHz)	Source of energetic particles	Negative pressure threshold (bars)	Reference
Bromoethane	Ultrasonic	30	47	Pu/Be-n	3.1	Greenspan and Tscheig 1967
"	"	31	36	"	3.1	"
Ethylene bromide	Spinner	20	n.a.	$^{210}\text{Po}-\alpha$	22.19	Hahn 1961
Acetophenone	"	≈ 20.5	n.a.	$^{210}\text{Po}-\alpha$	33.23	Hahn 1961
Cyclohexanol	"	≈ 23.45	n.a.	$^{210}\text{Po}-\alpha$	48.74	Hahn 1961
Methyl iodide	"	20	n.a.	$^{137}\text{Cs}-\gamma$	50	Hahn 1961
Freon 12	Ultrasonic				?	Hahn and Peacock 1963
Freon 115	"				?	Hahn and Peacock 1963
Methyl acetate	"			U/fission	?	Hahn and Peacock 1963
1-propanol		27				Greenspan and Tscheig 1967
1-propanol	"	29		Th(fast fission)	-10db ^a	Greenspan and Tscheig 1967
Water	"	?	?	U/n fission	10-20	Greenspan and Tscheig 1967
1-butanol	"	26	40	Pu/Be-n	8.6	Greenspan and Tscheig 1967
Carbon tetrachloride	"	24	50	Pu/Be-n	4.9	Greenspan and Tscheig 1967

^aApproximate. Relative to the threshold for Pu/Be-n induced cavitation.

APPENDIX B

EFFECT OF SURFACE CURVATURE ON ENTHALPY OF EVAPORATION

APPENDIX B. EFFECT OF SURFACE CURVATURE ON ENTHALPY OF EVAPORATION

Not only is the equilibrium vapor pressure higher over the curved surface of a small droplet, compared with a plane surface, but the enthalpy of evaporation is reduced. If the same curvature effects do take place inside a bubble, with the opposite sign, then the enthalpy of evaporation would be increased. However, it seems that the quantitative change would be very small.

Using the formula 8.48 in Dufour and Defay (1963), with a change of nomenclature and substituting $1/\rho_\ell$ for the specific volume of the liquid, one may write, for a concave surface of radius r ,

$$H_{vr} = H_{v\infty} + 2\sigma/(\rho_\ell r) \quad (\text{B.1})$$

Enthalpy of evaporation from a surface of radius r = enthalpy of evaporation from a plane surface + $2\sigma/(\rho_\ell r)$.

By inspection, we can see that the effect will be larger for liquids with a high value of $2\sigma/r$, i.e., in those cases where the ΔP between the bubble interior and the liquid is greatest. Therefore, as an extreme example, take the case of the cyclohexanol results reported in Tables 2 and 3.

$$\begin{aligned} H_{vr_{crit}} - H_{v\infty} &= \frac{2\sigma}{r_{crit} \rho_\ell} \\ &= \frac{2 \times 33.58 \times 10^{-3}}{13.78 \times 10^{-9} \times 9.605 \times 10^3} \\ &= 5.1 \times 10^2 \end{aligned}$$

The correction is only 0.5 kJ/mol compared with an $H_{v\infty}$ value of 61 kJ/mol, a correction of less than 1%. In other cases listed in Table 2, the correction is even smaller. This effect has been ignored in calculating the W_v energy term.

APPENDIX C

CALCULATIONS ON RIEPE AND HAHN'S BUBBLE CHAMBER EXPERIMENT RESULTS AND COMPARISONS WITH THEIR CALCULATIONS

APPENDIX C. CALCULATIONS ON RIEPE AND HAHN'S BUBBLE CHAMBER EXPERIMENT RESULTS AND COMPARISONS WITH THEIR CALCULATIONS

As a check of my own methods, I have recalculated the critical radii and the energy terms using modern values of the liquid and vapor properties (Yaws 1996). I do not know how Riepe and Hahn solved the two nasty simultaneous Eqs. (3) and (4) for the critical radius, but by their standards I probably had it easy, using the automatic iterative solution finder in TK Solver.

This close to the critical temperature, the vapor density used in Eq. (5b) is significantly different from the ideal gas value P/RT . Under the conditions shown in Table 1, the vapor densities given by Yaws are 15–20% higher than the ideal gas calculation.

First, from Eq. (4) of the main text,

$$P_{vrcrit} = P_{voo} \exp \left[-2\sigma / (RT\rho_l r_{crit}) \right] , \quad (C.1)$$

and for convenience write

$$P_{vrcrit} = \alpha \times P_{voo} . \quad (C.2)$$

Then, by inspection of Eq. (C.1),

$$\alpha = \exp \left[-2\sigma / (RT\rho_l r_{crit}) \right] , \quad (C.3)$$

which allows one to rewrite Eq. (3) from the main text of this report as

$$r_{crit} = 2\sigma / (\alpha \times P_{voo} - P_{ext}) , \quad (C.4)$$

and in the Riepe and Hahn experiments, P_{ext} is equal to the initial (i.e., vapor) pressure of the chamber minus the measured pressure drop, ΔP , resulting from the expansion (i.e., ΔP is the pressure drop measured by Riepe and Hahn). Therefore,

$$r_{crit} = 2\sigma / [\alpha \times P_{voo} - (P_{voo} - \Delta P)]$$

or

$$r_{crit} = 2\sigma / [\Delta P - (1 - \alpha)P_{voo}] . \quad (C.5)$$

Given a reasonable guess, or estimate, as a starting point, TK Solver can iteratively solve the two simultaneous Eqs. (C.3) and (C.5) for r_{crit} and α . I normally used $\alpha = 1.0$ as that guess. Once α and r_{crit} are known, P_{vrcrit} , the vapor pressure expected inside the critical bubble, is easily evaluated from Eq. (C.2). The vapor density can then be obtained from the Yaws data base. Table C.1 summarizes the results for the two liquids shown in Table 1 of the main text.

The actual TK Solver models and the input data are shown at the end of this appendix. The results are summarized in Table C.1, which also shows, in the last two lines, the difference between the Yaws value for the vapor density and the ideal gas value, P/RT , at the same temperature and pressure.

In a separate calculation, one can then compute the vapor content of the bubble and evaluate Eqs. (5a) through (5d) for comparison with Table 1. The properties needed for these computations are shown in Table C.2: I have converted the Yaws data to SI units where necessary, using the conversion factors in ASTM (1992).

Table C.1. Calculated parameters of critical bubbles

<i>Material</i>		
Name	Freon 12	Propane
Composition	CCl_2F_2	C_3H_8
<i>Conditions</i>		
Temperature, °C	29.4	18.5
Measured ΔP , Pa	200×10^3	197×10^3
<i>Properties (Yaws)^a</i>		
Molecular weight	120.913	44.096
Surface tension, N/m	8.23×10^{-3}	7.78×10^{-3}
Liquid density, mols/m^3	10.680×10^3	11.407×10^3
Vapor pressure, ^b Pa	722.6×10^3	806.5×10^3
<i>Calculated values</i>		
α	0.9928	0.9931
r_{crit} , m	84.52×10^{-9}	81.28×10^{-9}
P_{vrcrit} , ^c Pa	721.35×10^3	800.94×10^3
Vapor density, ^d mols/m^3	336.1	373.4
P_{vrcrit}/RT , mols/m^3	286.8	330.3

^aConverted by this author to SI units where necessary, using the factors in ASTM (1992).

^bOver a plane surface.

^cOver a concave spherical surface of radius r_{crit} .

^dAt the temperature shown and pressure P_{vrcrit} .

Table C.2. Properties of Freon 12 and propane

<i>Material</i>		
Name	Freon 12	Propane
Composition	CCl_2F_2	C_3H_8
<i>Conditions</i>		
Temperature T , °C	29.4	18.5
Measured ΔP , ^a Pa	200×10^3	197×10^3
P_{ext} ^b	522.6×10^3	609.5×10^3
<i>Vapor properties (Yaws)</i>		
Pressure P_{vap} ^c Pa	722.6×10^3	806.5×10^3
Pressure P_{vrcrit} ^d Pa	721.35×10^3	800.94×10^3
Density, ^e mols/m ³	336.1	373.4
<i>Liquid properties (Yaws)</i>		
Surface tension, N/m	8.23×10^{-3}	7.78×10^{-3}
Density, mols/m ³	10.680×10^3	11.407×10^3
Enthalpy of evap., J/mol	16.54×10^3	15.25×10^3
Specific heat, J/mol · K	122.70	115.24
Thermal conductivity, W/m · K	0.0731	0.1627
Viscosity, Pa · s	0.255×10^{-3}	0.107×10^{-3}
Thermal diffusivity ^f	55.8×10^{-9}	123.7×10^{-9}

^aFor ^{210}Po recoils.

^b $P_{\text{vap}} - \Delta P$.

^cOver a plane surface.

^dVapor pressure over the concave curved surface of the critical bubble—see Table C.1.

^eAt the temperature listed (T) and a pressure of P_{vrcrit} .

^fCalculated as thermal conductivity/(density $\times$ specific heat).

The two calculation steps could easily be combined into a single TK model, but for flexibility and ease of cross-checking, I chose not to do that.

Using these values of the properties and the values of r_{crit} shown in Table C.1, the vapor content of the bubble, the speed of expansion, and the various energy terms are recalculated and compared with Riepe and Hahn's results in Table C.3.

The present calculations of overall bubble nucleation center formation energy are within a few percent of the Riepe and Hahn results. The only significant disagreement, and that only in percentage terms, is in the small W_5 term for propane, and for that term Riepe and Hahn explicitly write that they had no precise data on thermal conductivity or viscosity.

Table C.3. ^{210}Po recoils

Liquid temperature	CCl_2F_2 (29.4°C)		C_3H_8 (18.5°C)	
	Riepe and Hahn	West	Riepe and Hahn	West
Calculated critical radius, Å	861	845	829	813
Growth velocity, cm/s	304	264	610	609
No. of molecules in critical bubble	5.55×10^5	5.12×10^5	5.52×10^5	5.06×10^5
Surface energy, W_{s1} , keV	4.9	4.6	4.3	4.0
PV work, W_e , keV	8.7	8.2	8.9	8.6
Latent heat, W_v , keV	94.7	87.8	89.0	80.0
Kinetic energy, W_4 , keV	0.3	0.2	~0.4	0.4
Viscous work, W_5 , keV	1.8	1.5	~2.6	1.4
Total energy, keV	110.4	102.3	105.2	94.4

Riepe and Hahn's results and calculations showed that Seitz' thermal spike theory of bubble chamber operation is in quantitative agreement with the observed boiling nucleation behavior of α -recoils (i.e., of heavy, slow moving energetic particles).

Sta	Input	Name	Output	Unit	Comment
					CCl2F2 @ 29.4deg C.2-21-97.
	8.3145	R		J/mol.K	Molar gas constant.
	302.55	T		K	Liquid temperature.
	.00823	sigma		N/m	Surface tension.
	10680	rhoL		moles/m^3	Liquid density.
	726600	Pv		Pa	Vapor pressure over a plane interface.
	200000	deltaP		Pa	Pressure drop below Pv.
		alpha	.992777225		Ratio of vapor pressures over curved & flat surfaces
		rcrit	8.45178E-8	m	Critical radius taking account of curvature.
		Pvr	721351.932	Pa	Vapor pressure taking account of curvature.

Variable Sheet

R&H.TKW

Page 1

St Rule

alpha=exp(-2*sigma/(R*T*rhoL*2*sigma/(deltaP-(1-alpha)*Pv)))
rcrit=2*sigma/(deltaP-(1-alpha)*Pv)
Pvr=alpha*Pv

Rule Sheet

R&H.TKW

Page 1

Sta	Input	Name	Output	Unit	Comment
					C3H8 @ 18.5deg C.2-21-97.
	8.3145	R		J/mol.K	Molar gas constant.
	291.65	T		K	Liquid temperature.
	.00778	sigma		N/m	Surface tension.
	11407	rho1		mols/m^3	Liquid density.
	806500	Pv		Pa	Vapor pressure over a plane interface.
	197000	deltaP		Pa	Pressure drop below Pv.
		alpha	.993103071		Ratio of vapor pressures over curved & flat surfaces
		rcrit	8.12797E-8	m	Critical radius taking account of curvature.
		Pvr	800937.627	Pa	Vapor pressure taking account of curvature.

Variable Sheet

R&H.TKW

Page 1

St Rule

alpha=exp(-2*sigma/(R*T*rho1*2*sigma/(deltaP-(1-alpha)*Pv)))
rcrit=2*sigma/(deltaP-(1-alpha)*Pv)
Pvr=alpha*Pv

Rule Sheet

R&H.TKW

Page 1

Sta	Input	Name	Output	Unit	Comment
					CCl2F2 @ 29.4 C
	120.913	M			Molecular weight
	522600	Pext		Pa	Pvap-deltaP
	8.452E-8	rcrit		m	
	.00823	sigma		N/m	
	10680	rho1		mols/m^3	
	16540	Hv		J/mol	
	122.7	spht		J/mol.k	
	.0731	lambda		W/m.k	
	.000255	eta		Pa.s	
	336.1	rho vr		mols/m^3	
		D	5.5783E-8		
		Wsl	7.388E-16	J	
		We	1.3217E-15	J	
		Wv	1.406E-14	J	
		W4	3.4143E-17	J	
		W5	2.4173E-16	J	

Variable Sheet

RH2.TKW

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St Rule

D=	$\lambda / (\rho_1 \cdot \text{spht})$
Wsl=	$4 \cdot \pi \cdot (\text{rcrit})^2 \cdot \sigma$
We=	$(4/3) \cdot \pi \cdot (\text{rcrit})^3 \cdot P_{\text{ext}}$
Wv=	$(4/3) \cdot \pi \cdot (\text{rcrit})^3 \cdot \rho_{\text{vr}} \cdot H_v$
W4=	$32 \cdot \pi \cdot D^2 \cdot \rho_1 \cdot \text{rcrit} \cdot (M/1000)$
W5=	$64 \cdot \pi \cdot \eta \cdot D \cdot \text{rcrit}$

Rule Sheet

RH2.TKW

Page 1

Sta	Input	Name	Output	Unit	Comment
					C3H8 @ 18.5 C
	44.096	M			Molecular weight
	609500	Pext		Pa	Pvap-deltaP
	8.128E-8	rcrit		m	
	.00778	sigma		N/m	
	11407	rho1		mols/m^3	
	15250	Hv		J/mol	
	115.24	spht		J/mol.k	
	.1627	lambda		W/m.k	
	.000107	eta		Pa.s	
	373.4	rhovr		mols/m^3	
		D	1.23769E-7		
		Wsl	6.4589E-16	J	
		We	1.3709E-15	J	
		Wv	1.2808E-14	J	
		W4	6.2962E-17	J	
		W5	2.1643E-16	J	

Variable Sheet

RH2.TKW

Page 1

St Rule

D=	$\lambda / (\rho_1 \cdot \text{spht})$
Wsl=	$4 \cdot \pi \cdot \text{rcrit}^2 \cdot \sigma$
We=	$(4/3) \cdot \pi \cdot \text{rcrit}^3 \cdot P_{\text{ext}}$
Wv=	$(4/3) \cdot \pi \cdot \text{rcrit}^3 \cdot \rho_{\text{avr}} \cdot H_v$
W4=	$32 \cdot \pi \cdot D^2 \cdot \rho_1 \cdot \text{rcrit} \cdot (M/1000)$
W5=	$64 \cdot \pi \cdot \eta \cdot D \cdot \text{rcrit}$

Rule Sheet

RH2.TKW

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APPENDIX D

AN ADDITIONAL SURFACE ENERGY TERM?

APPENDIX D. AN ADDITIONAL SURFACE ENERGY TERM?

Bugg (1959) introduced an additional surface energy term. This was also reported on by Tenner (1963), Hugentobler et al (1963), and Peyrou (1967). Using Peyrou's notation, there is an additional surface energy, W_{s2} , needed to account for the extra internal energy needed by the vapor in the bubble in order to form the surface in an adiabatic way (i.e., without input from anything but the energy of the vapor).

$$W_{s2} = -4\pi r^2 T \partial \sigma / \partial T . \quad (D.1)$$

At least in most cases, $-T \partial \sigma / \partial T$ is greater than σ , so Bugg's term would be a large part of the surface energy.

In the Yaws data base, surface tension at temperature T Kelvin is represented by a correlation of this form:

$$\sigma(T) = (A + BT)^n ; \quad (D.2)$$

therefore,

$$T \frac{\partial \sigma}{\partial T} = nBT(A + BT)^{n-1} , \quad (D.3)$$

and it is easy to compute the value of W_{s2} , using the values of A , B , and n from Yaws. Between the melting point and the critical point, T is always positive and $\partial \sigma / \partial T$ negative, so that $-T \partial \sigma / \partial T$ is positive and adds to the surface energy. Table D.1 is an extension of the West figures and the West columns in Table C.3 to include the W_{s2} energy: one does not know values of σ , to say nothing of $\partial \sigma / \partial T$, Riepe and Hahn would have used if they were to have calculated this term, and so no comparison with their results is possible.

The calculated total energy is still satisfactorily close to the recoil and alpha energy from the ^{210}Po decay, although it must be said that the addition of the W_{s2} term has not improved the agreement.

Of the $T \partial \sigma / \partial T$ term, Peyrou (1967) wrote, "The author, not being a specialist in thermodynamics, must confess that he is somewhat puzzled by this term."*

*This sentence in Peyrou's 1967 paper was footnoted as follows: "Since this paper was written, it has become clear to the author that, indeed, the adiabaticity of the process inside the bubble has nothing to do with the adiabaticity of surface formation; that is why extra energy is needed to take this fact into account. However, since heat diffusion is very fast around a little spherical surface (see Section III.D) it is still not clear that surface formation is indeed an adiabatic process."

Table D.1. Extension of West figures and columns in Table C.3

Temperature	CCl ₂ F	C ₃ H ₈
	29.4°C/302.55 K	18.5°C/291.65 K
<i>A</i> (from Yaws)	24.6498	26.4582
<i>B</i> (from Yaws)	-0.06403	-0.07154
<i>n</i> (from Yaws)	1.267	1.192
σ (from Yaws), N/m	8.23	7.78
$T\partial\sigma/\partial T$, N/m	-38.37	-34.61
W_{s1} from Table C.3, keV	4.6	4.0
W_{s2} from Eqs. (C.1) and (C.3), keV	21.4	17.8
W_e from Table C.3, keV	8.2	8.6
W_v from Table C.3, keV	87.8	80.0
W_4 from Table C.3, keV	0.2	0.4
W_5 from Table C.3, keV	1.5	1.4
Total energy, keV	123.7	112.2

APPENDIX E

FLUID PROPERTIES FOR EVALUATIONS OF ENERGY TERMS AND PRESSURE THRESHOLD FOR HAHN'S SPINNER EXPERIMENTS

APPENDIX E: FLUID PROPERTIES FOR EVALUATIONS OF ENERGY TERMS AND PRESSURE THRESHOLD FOR HAHN'S SPINNER EXPERIMENTS

The properties of the fluids in Hahn's spinner experiments were obtained from the Yaws database (Yaws 1996) and converted as necessary to the units used in this report. In a few cases (e.g., the viscosity of liquid acetophenone and cyclohexanol), the correlations used by Yaws were extrapolated a few degrees outside their applicable temperature range.

The data listed in Table E.1 are rounded to fit in the available space. The properties calculated from Yaws, such as molar densities, carried more significant figures in the TK solver models than in the table.

Table E.1. Liquid and vapor properties; 10-5-98

	1	2	3	4	5	6	7	8	9
Formula	'C5H12	'C2Cl3F3	'C2Cl3F3	'C6H14	'C3H6O	'C2HCl3	'C2H4Br2	'C8H8O	'C6H12O
Erecoil,keV	101	166	101	101	101	101	101	101	101
Erecoil,fJ	16.182	26.596	16.182	16.182	16.182	16.182	16.182	16.182	16.182
Zp	82	82	82	82	82	82	82	82	82
Tamb,K	293.15	293.15	293.15	293.15	293.15	293.15	293.15	293.65	296.60
Pcav(exp),bars	3.34	4.49	5.21	6.89	8.31	11.75	22.19	33.23	48.74
From Yaws data base									
Mwt	72.150	187.375	187.375	86.177	58.080	131.388	187.862	120.151	100.161
Zl	42	90	90	50	32	64	86	64	56
rhoI,kg/m^3	621	1576	1576	661	791	1466	2179	1028	962
sigma,mN/m	14.99	17.72	17.72	18.54	23.66	29.40	38.82	39.46	33.58
TdsigdT,mN/m	-31.73	-33.16	-33.16	-32.56	-36.26	-38.88	-39.09	-33.09	-31.41
Cpliq,J/mol.K	157.03	168.26	168.26	191.40	126.66	123.05	131.03	199.31	217.80
eta,mPa.s	.225	.734	.734	.310	.322	.571	1.749	1.891	29.540
Kliq,w/m.K	1.16E-1	7.71E-2	7.71E-2	1.23E-1	1.63E-1	1.18E-1	1.01E-1	1.75E-1	1.34E-1
Pcrit,bars	33.81	34.15	34.15	30.12	47.02	49.10	54.77	38.40	37.49
Tcrit,K	460.43	487.25	487.25	507.43	508.20	571.00	650.15	701.00	625.15
rhoCrit,kg/m^3	235.9	576.0	576.0	233.0	277.9	513.2	718.2	319.6	311.1
Hca,Btu/lb	237.18	103.24	103.24	295.78	295.34	154.14	146.18	403.20	429.29
Pvap,mmHg	575.25	270.75	270.75	122.39	184.35	58.16	10.18	0.28	0.66
Hvap,kJ/mol	25.27	28.86	28.86	32.28	32.56	35.27	38.38	53.89	61.09
Calc. from Yaws									
rhoI,kmol/m^3	8.607	8.411	8.411	7.670	13.619	11.158	11.599	8.556	9.605
rhoCrit,kmol/m^3	3.270	3.074	3.074	2.704	4.785	3.906	3.823	2.660	3.106
Hca,kJ/mol	39.78	44.97	44.97	59.25	39.87	47.07	63.83	112.61	99.95
D,m^2/s	8.553E-8	5.448E-8	5.448E-8	8.344E-8	9.449E-8	8.565E-8	6.665E-8	1.023E-7	6.425E-8
Pvap,bars	7.67E-1	3.61E-1	3.61E-1	1.63E-1	2.46E-1	7.75E-2	1.36E-2	3.73E-4	8.80E-4
sigeff,mN/m	46.72	50.88	50.88	51.10	59.92	68.28	77.91	72.55	64.99
etaeff,mPa.s	2.383E-1	7.555E-1	7.555E-1	3.238E-1	3.407E-1	6.024E-1	1.785	1.917	29.555

INT TABLE: Properties

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